OPTIMIZING TECHNOLOGY TO REDUCE MERCURY AND ACID GAS EMISSIONS FROM ELECTRIC POWER PLANTS

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Principal Authors **Jeffrey C. Quick**
David E. Tabet
Sharon Wakefield
Roger L. Bon

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ABSTRACT

County-average hydrogen values are calculated for the part 2, 1999 Information Collection Request (ICR) coal-quality data, published by the U.S. Environmental Protection Agency. These data are used together with estimated, county-average moisture values to calculate average net heating values for coal produced in U.S. counties. Finally, 10 draft maps of the contiguous U.S. showing the potential uncontrolled sulfur, chlorine and mercury emissions of coal by U.S. county-of-origin, as well as expected mercury emissions calculated for existing emission control technologies, are presented and discussed.

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INTRODUCTION

Background

Switching to low-mercury-emission coal may be an effective strategy to comply with impending regulations that are intended to reduce mercury emissions from electric utilities. For example, despite proven emission control technology, burning low-sulfur coal is the most popular method to reduce sulfur emissions. Because technology to reduce mercury emissions is considerably less certain, burning low-mercury coal is a likely method to reduce mercury emissions. Like sulfur, the amount of mercury in U.S. coal shows substantial geographic variation. Furthermore, mercury emissions from similar types of power plants are largely correlated with the amount of mercury in the coal. However, unlike sulfur, mercury emissions also vary with the abundance of other elements in the coal such as chlorine and sulfur, which influence mercury capture by emission control technologies. Consequently, mercury emission factors vary according to the relative abundance of several elements in the coal, and are specific to different emission control technologies.

This project is using Geographic Information System technology (ArcView GIS) to create detailed maps to show where U.S. coal with low mercury and acid-gas emissions might be found. The map series will show geographic variation of mercury, chlorine, and sulfur in coal, as well as the mercury emission penalty calculated for data aggregated by U.S. county-of-origin using equations specific to power plants classified by boiler type and flue gas emission controls. Removing mercury from flue gas is a technically complex task – different technologies will be required for different coals. Maps showing the geographic variation of mercury and acid gas emission factors for U.S. coal will help locate the best coal for each technology and identify the best technology for each coal.

Coal quality data used in this study were described in a previous report (Quick and others, 2004). Briefly, these data were selected from five data sets and include: 19,507 FERC 423 data records (USEIA, 2003a), 25,818 ICR data records (USEPA, 2003), 5,602 CTRDB data records (USEIA, 2003b), 5,045 COALQUAL data records (Bragg and others, 1997), and 73 PSU-DOE data records (Anonymous, 1990; Davis and Glick, 1993; Scaroni and others, 1999).

Recent Developments

The U.S. Environmental Protection Agency (USEPA) is expected to issue new rules limiting mercury emissions from coal-fired electric power plants by March 15, 2005, with enforcement beginning as early as April 2008. The proposed rules are described in the USEPA (2004) federal register notice; two different options are described in the notice.

The Maximum Achievable Control Technology (MACT) option would require each power plant to limit mercury emissions according to the rank of the coal burned, with special limits for plants burning waste coal (refuse) or using integrated, combined-cycle technology (IGCC). The proposed MACT emission limits are listed in table 1.

Table 1. Proposed MACT mercury emission limits (section 112, MACT rule option) for existing and new coal-fired electric utility steam generating units (USEPA, 2004, tables 1 and 2, pages 4662 and 4663).

		Input based limit pounds Hg per trillion gross Btu of coal		Output based limit pounds Hg x 10^{-6} per gross MWh manufactured
Existing Plants	bituminous	2.0	or	21
	subbituminous	5.8	or	61
	lignite	9.2	or	98
	IGCC	19.0	or	200
	refuse	0.38	or	4.1
New Plants	bituminous			6
	subbituminous			20
	lignite			62
	IGCC			20
	refuse			1.1

The cap-and-trade option would limit total mercury emissions from all power plants to a maximum 15 tons per year by 2018. Each power plant would be required to have mercury emission allowances sufficient to equal its annual mercury emissions. The allowances would be distributed by state or federal administrators, and could be used, saved, purchased, or sold. The USEPA (2004) cap-and-trade proposal allocates allowances to U.S. States according to their proportional share of coal energy consumption, modified by the rank of the coal consumed. A state's fractional share of the proposed 15-ton cap would be calculated as its average coal energy consumption (highest annual average for 3 of 4 years between 1998 and 2002, of the summed energy content of coal burned in electric utilities), multiplied by a factor of 1 for bituminous, 1.25 for subbituminous, and 3 for lignite rank coal, and finally divided by the sum of the results calculated for all 50 states. Additionally, under the cap-and-trade rule, newly constructed power plants would need to meet the same standards as those listed in table 1 for new plants under the proposed MACT rule. Although the form of the final rule remains uncertain, the proposed emission limits shown in table 1 are useful benchmarks to evaluate the geographic variation of potential mercury emissions.

Scope of This Report

This report describes the progress made during the second six months of this 24-month project. Results of tasks 4, 5, and 6 (figure 1) are described and discussed.
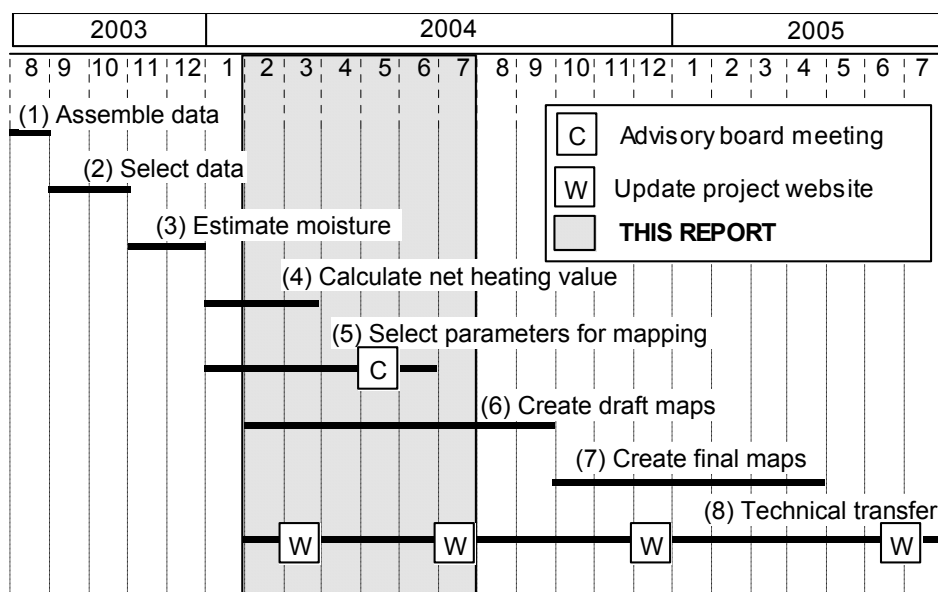
Figure 1. Schedule of project tasks.

EXECUTIVE SUMMARY

Draft maps showing the geographic variation of mercury and acid gas emission factors for U.S. coals were constructed using coal assay data aggregated by U.S. county-of-origin. Specific tasks accomplished during the second six months of this two-year project include:

- Coal hydrogen values were estimated for the ICR data using equations based on selected COALQUAL data records.

- Net coal heating values were calculated for the ICR data by U.S. county-of-origin.

- Published emission factors that predict mercury capture for power plants classified by air pollution controls were selected and applied to the ICR data.

- Draft maps were made using ICR data aggregated by county-of-origin. The maps show potential uncontrolled mercury, sulfur, and chlorine emissions, as well as predicted mercury emissions from coal burned in power plants classified by air pollution controls.

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- High-mercury coal is produced in parts of Oklahoma, Texas, Ohio, Pennsylvania, Kentucky, Alabama, and Tennessee, whereas low-mercury coal is common in the western U.S., Eastern Interior Province, and the Central Appalachian Region.

- Coal from the Northern Appalachian Region (Ohio and parts of Pennsylvania) has notably high mercury concentrations compared to U.S. coal produced elsewhere.

- Much subbituminous and some lignite coal should comply with the proposed MACT rule using existing technology. Bituminous compliance coal for power plants with Electrostatic Precipitator (ESP) controls is rare. Plants equipped with Flue Gas Desulfurization (FGD) controls may find bituminous compliance coal in some western U.S. counties, the Eastern Interior Province, and the Central Appalachian Region. With notable exceptions (for example, numerous counties in Ohio and the western U.S.), fabric filters may be an effective technology for bituminous coal.

EXPERIMENTAL

The proposed MACT rule includes both input-based (pounds Hg per trillion Btu) and output-based (pounds Hg x 10[-6] per megawatt-hour electricity manufactured) emission limits (table 1). The output-based limits assume 32 percent efficiency (10,667 gross Btu/kilowatt-hour) for existing power plants and 35 percent efficiency (9,833 gross Btu/kilowatt-hour) for new power plants (USEPA, 2004). Although the USEPA used the gross heating value of coal[1] to calculate the output-based emission limits (Cole, 2003), figure 2 shows that output-based emissions are better calculated from fuel emission factors expressed on a net energy basis. Accordingly, we use emission factors expressed on a net energy basis to calculate output-based emissions. This required that we calculate county-average, ICR net heating values.
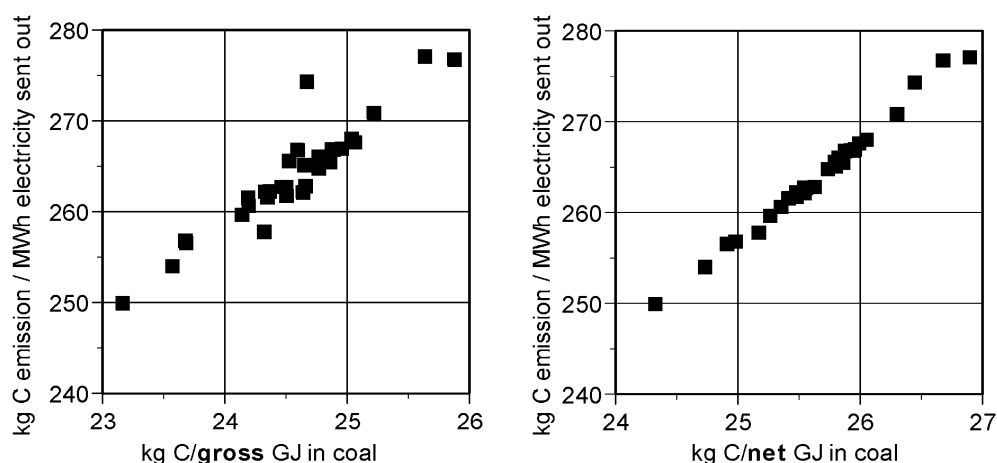
Figure 2. Emissions expressed on an output basis (vertical axes) are better estimated if the fuel emission factor is expressed on a net energy basis (right plot) rather than on a gross energy basis (left plot). Data show output-based carbon emissions calculated by Juniper (1998) for commercial coals in a model 500 MW plant equipped with ESP and FGD emissions controls.

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[1] The gross coal heating value, (also called the higher heating value) is the familiar Btu/lb (or MJ/kg) value reported from the laboratory. The gross heating value is measured by using a high-pressure, constant-volume combustion bomb. Because water vapor from combustion condenses inside the combustion bomb the gross heating value includes the latent heat of water vapor. Unlike the laboratory combustion bomb, combustion in a coal-fired boiler occurs at constant pressure and moisture from combustion exits the boiler with the flue gas. Consequently, the net heating value (also called the lower heating value) does not include the latent heat of water vapor and is a better measure of the energy available to the boiler than the gross heating value.

Calculation of the Net Heating Value (Task 4)

The net heating value is calculated as:

$$Btu_{net} = Btu_{gross} - 92.7(0.1119M + H) \qquad\qquad (1)$$

where: Btu_{gross} is the familiar Btu per pound value reported from the laboratory and expressed on a moist, whole-coal basis,

M is the weight percent moisture content of the coal,

H is the weight percent hydrogen of the coal (not including hydrogen in coal moisture) expressed on a moist, whole-coal basis,

0.1119 is the gravimetric factor applied to the moisture value (M) to obtain the weight percent hydrogen in coal moisture and,

92.7 is the Btu penalty, which is largely due to the latent heat of water vapor, which is lost from the boiler with the combustion flue gas.

Note that the ICR data do not include moisture or hydrogen values, which are required for equation 1. County-average, ICR moisture values were estimated in an earlier report (Quick and others, 2004). County-average ICR hydrogen values were calculated using predictive equations obtained by regression analysis, which is described below.

Predicting ICR Coal Hydrogen Values

A multivariate regression method was applied to selected COALQUAL data (Mott-Spooner values within ±250 Btu) to develop a set of geographically specific equations to predict coal hydrogen content using dry-basis Btu/lb, ash, and sulfur values. The equations were validated using the PSU-DOE data, and used to estimate ICR coal hydrogen values.

The dependent COALQUAL variable was dry-basis hydrogen. Note that moist-basis hydrogen values, which include hydrogen in coal moisture, are listed in the COALQUAL data

set. Consequently, the COALQUAL hydrogen values were adjusted to a dry basis by subtracting

the stochiometric contribution of water to hydrogen (0.1119 x moisture), and multiplying the

result by $\dfrac{100}{100 - moisture}$ (ASTM, 2000a).

The four independent variables used in the regression analysis (Btu_{dmmf}, Btu_{dmmf}^2, $MM_{Parr, dry}$,

and lbs S/million Btu) were calculated for the selected COALQUAL data records using the

equations:

$$Btu_{dmmf} = \frac{100 \times \left(Btu_{dry} - 50S_{dry}\right)}{100 - (1.08Ash_{dry} + 0.55S_{dry})} \qquad (2)$$

$$Btu_{dmmf}^2 = Btu_{dmmf} \times Btu_{dmmf} \qquad (3)$$

$$MM_{Parr,dry} = 1.08Ash_{dry} + 0.55S_{dry} \qquad (4)$$

$$lbs\ S/million\ Btu = \frac{10^6}{Btu_{dry}} \times \frac{S_{dry}}{100} \qquad (5)$$

where, Btu_{dry} is the dry-basis Btu per pound value,

S_{dry} is the dry-basis weight percent sulfur, and

Ash_{dry} is the dry-basis weight percent ash.

Although the regression equations were obtained using relationships observed in the

COALQUAL data, they were used to predict ICR coal hydrogen values. Consequently, the

selection of the independent variables was necessarily constrained by the available ICR assay

data (Btu/lb, ash, S, Cl, Hg, and estimated moisture). Considering this constraint, the

independent variables were selected to indicate coal rank, (Btu_{dmmf} and Btu_{dmmf}^2), coal grade

($MM_{Parr,dry}$), and coal type (lbs S/million Btu), all of which may influence the hydrogen content of

coal. For example, the influence of coal rank is illustrated in figure 3, which shows that coal

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hydrogen increases slightly through the coalification series to reach a maximum in the high

volatile A bituminous stage, and then decreases as rank advances to the anthracite stage.
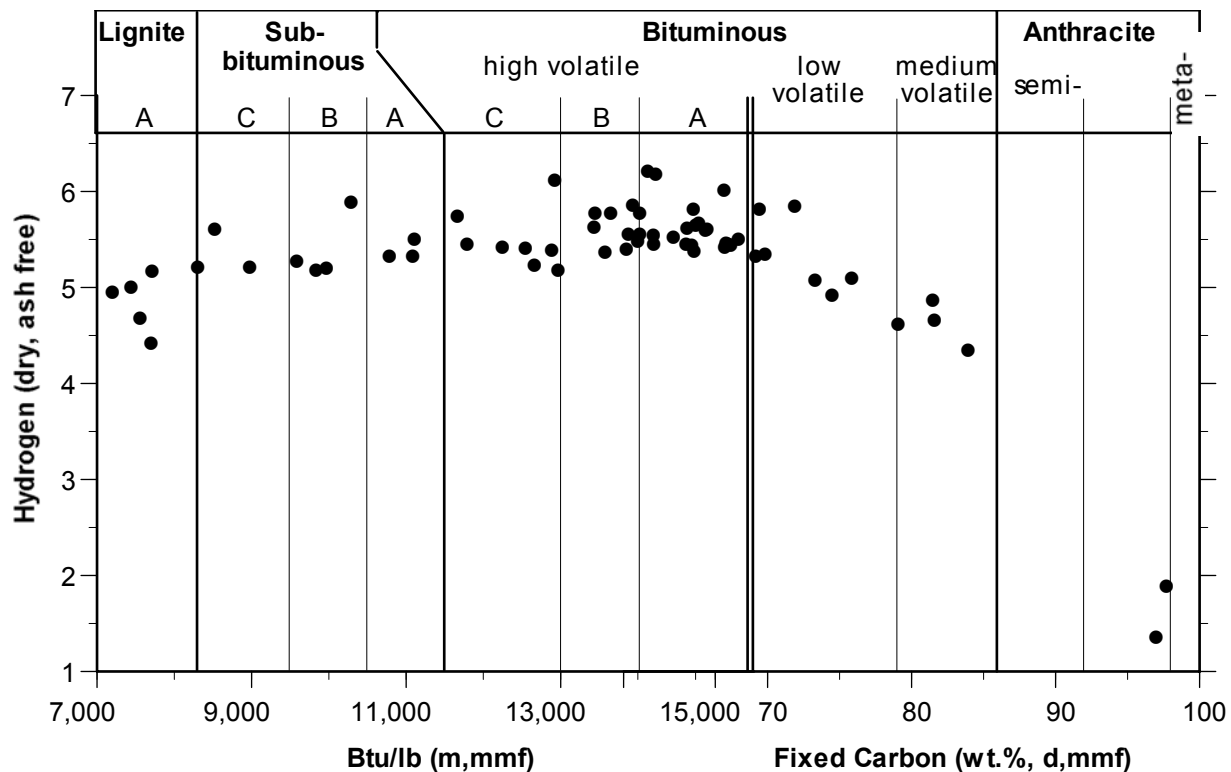
Figure 3. Variation of coal hydrogen with ASTM (1990) coal rank. Constructed using PSU-DOE data.

Note that the ASTM rank classification (figure 3) requires two parameters: (1) the Btu

value on a moist, mineral-matter-free basis [Btu/lb (m,mmf)] and (2) the fixed carbon value on a

dry, mineral-matter-free basis [Fixed Carbon (d,mmf)]. Regrettably, neither parameter could be

used as an independent variable in the regression analysis to predict coal hydrogen. We used the

Btu value on a dry, mineral-matter-free basis (instead of the moist, mineral-matter-free basis

used in the ASTM rank classification) because the COALQUAL moisture values are

systematically lower than those observed in other data sets (figure 4). The fixed carbon

parameter could not be used because fixed carbon values are not included in the ICR data.
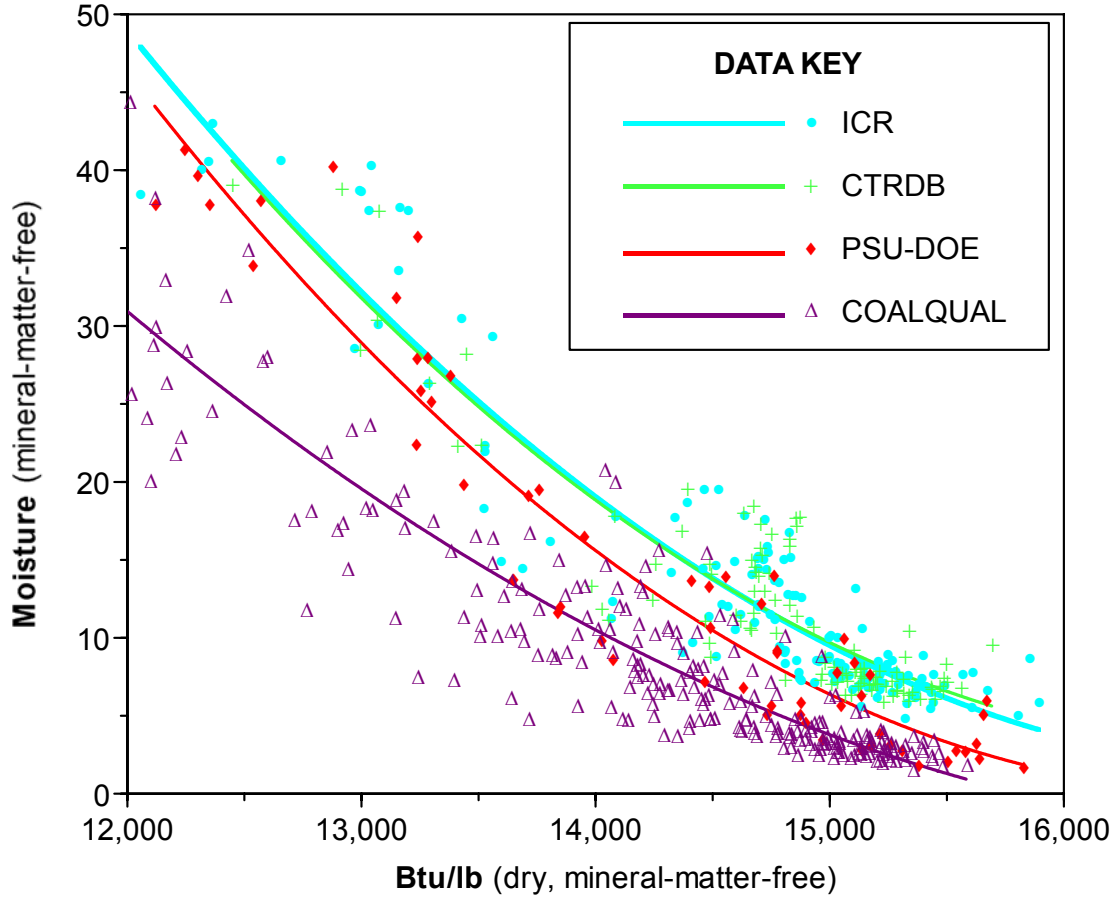
Figure 4. COALQUAL moisture values are lower than moisture values for other data sets. Notes: The ICR moisture values are estimated, county-average, as-shipped values. The CTRDB, and COALQUAL data points show measured, county-average moisture values (as-received basis). The PSU-DOE data points show equilibrium moisture values for single coal assays. Data from U.S. counties with medium volatile bituminous or higher rank coal are not shown. Also not shown are data for 46 counties included in the COALQUAL data where the average dry, mineral-matter-free Btu value is less than 12,000 (the ICR, CTRDB, and PSU-DOE data do not include data records where the dry, mineral-matter-free Btu/lb value is less than 12,000). The Btu/lb (dry, mineral-matter-free) values were calculated using equation 2 (see text). Moisture (mineral-matter-free) (M_{mmf}) was calculated as: M_{mmf} = Moisture [100/(1.08 Ash_{moist} + 0.55 $Sulfur_{moist}$)]. The best-fit lines correspond to: ICR $M_{mmf} = 1.82E^{-6}$ $Btu/lb_{d,mmf}^{2}$ - 6.22E^{-2} $Btu/lb_{d,mmf}$ + 533, (R^2 = 0.88); CTRDB $M_{mmf} = 1.92E^{-6}$ $Btu/lb_{d,mmf}^{2}$ - 6.47E^{-2} $Btu/lb_{d,mmf}$ + 549, (R^2 = 0.83); PSU-DOE $M_{mmf} = 2.05E^{-6}$ $Btu/lb_{d,mmf}^{2}$ - 6.88E^{-2} $Btu/lb_{d,mmf}$ + 576, (R^2 = 0.92); and COALQUAL $M_{mmf} = 1.17E^{-6}$ $Btu/lb_{d,mmf}^{2}$ - 4.05E^{-2} $Btu/lb_{d,mmf}$ + 349, (R^2 = 0.94).

The relationship between coal rank and coal hydrogen (figure 3) also shows that

hydrogen declines at higher ranks with increasing fixed carbon. The sharp decline of hydrogen

at high rank shown in figure 3 shows that different equations are required for high and low-rank

coals. Accordingly, fixed carbon values listed in the COALQUAL data were used to identify

U.S. counties with high-rank coal. COALQUAL data values from these counties (Btu_{dmmf},

$Btu_{dmmf}{}^2$, $MM_{Parr,dry}$, and lbs S/million Btu values) were used to establish equations to predict the

hydrogen content of high-rank coal, and the equations were applied to the ICR data originating

from the same counties.

Attempts to develop a single equation to predict hydrogen for high volatile A bituminous

(hvAb) and lower rank coals gave unsatisfactory results. The results overestimated coal

hydrogen in some geographic regions and underestimated coal hydrogen in others. For example,

the multiple regression equation based on all the COALQUAL data for hvAb and lower rank

coal, showed average residuals of -0.15% hydrogen for Western Interior coal and +0.23%

hydrogen for Gulf Coast coal. To avoid these systematic errors, equations to predict coal

hydrogen were determined for coal from each of the geographic regions shown in figure 5.

The regression equations used to predict coal hydrogen in this report are described in

table 2. Several results are noteworthy. Excluding high-rank coal, relatively large t-statistic

values, and consistently negative coefficients for the coal grade parameter ($MM_{Parr,dry}$) show the

strong influence of mineral matter content on coal hydrogen; coal hydrogen declines with

increasing mineral content. The general lack of significance (t-statistic <2) for the rank

parameter (Btu_{dmmf}) for coal from the Northern Great Plains and the High Rank groups may be

due to small range of variation of the Btu variable in coal from these areas. Although the type

parameter (lbs S/million Btu) is typically the least significant of the independent variables, its
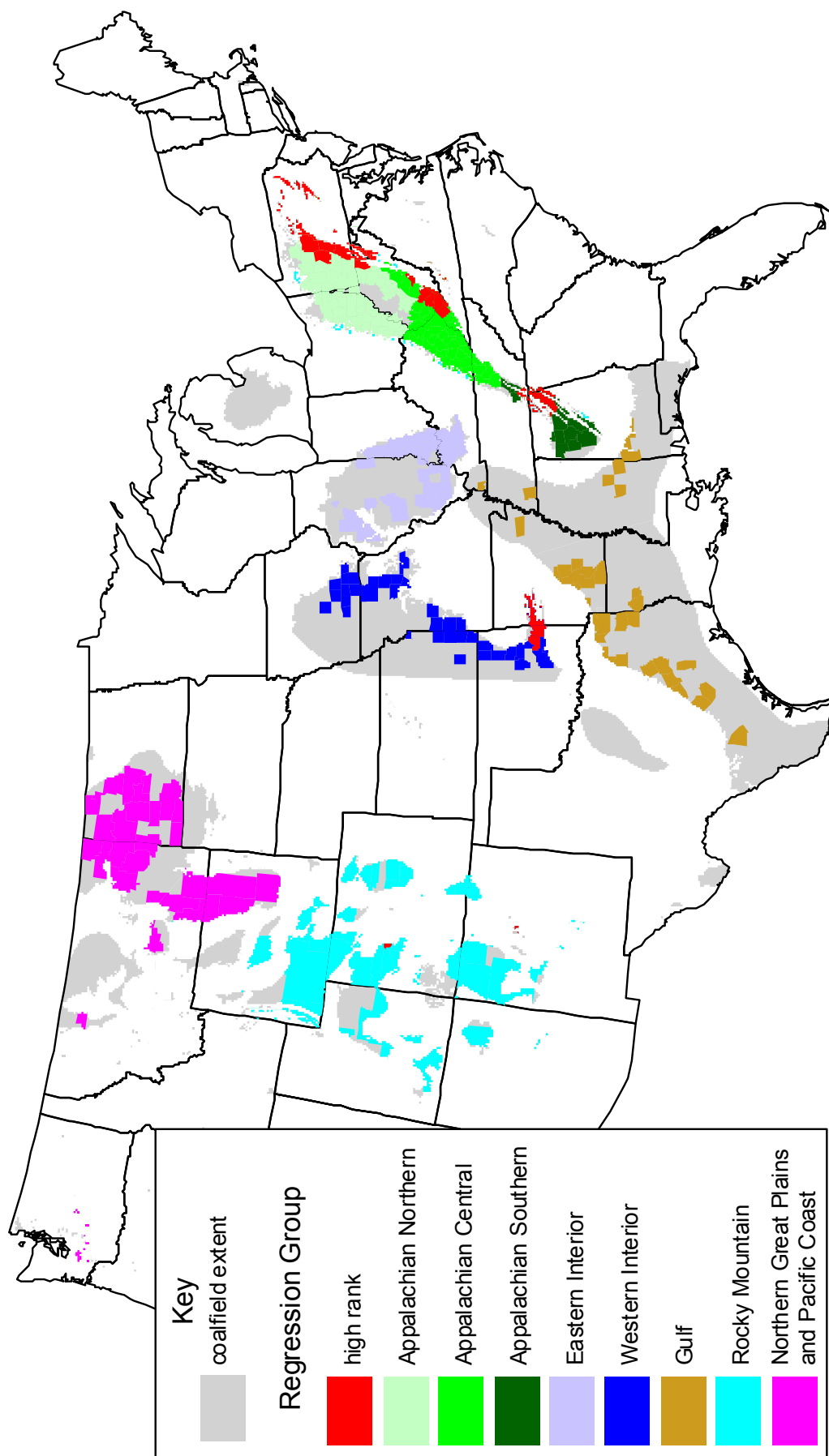
Figure 5. Regression equations to predict coal hydrogen content were developed for nine, coal-producing regional groups (modified from Trumbell, 1960).

Key

coalfield extent

Regression Group

high rank

Appalachian Northern

Appalachian Central

Appalachian Southern

Eastern Interior

Western Interior

Gulf

Rocky Mountain

Northern Great Plains and Pacific Coast

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generally positive coefficient is consistent with the geologic enrichment of coal hydrogen due to the preservation of otherwise labile hydrogen-rich compounds by an early diagenetic natural vulcanization process where aliphatic compounds are cross-linked by hydrogen sulfide from sulfate-reducing bacteria (Sinninghe Damste and others, 1989). The inability of the sulfur variable to predict coal hydrogen for coal from 5 of the 9 groups (t-statistic <2) is also noteworthy and may have varied origins; possibilities include (1) a late-stage abiogenic sulfide contribution to Western Interior coal (after diagenetic loss of labile hydrogen), (2) greater initial hydrogen of geologically younger (western U.S.) peat-forming biomass (more H-rich cellulose; Robinson, 1990) with early bacterial stripping of hydrogen by methanogenic bacteria, which thrive in the absence of dissolved sulfate (Belyaev and others, 1980), and (3) catagenetic loss of hydrogen associated with sulfur in aliphatic structures, as aliphatic sulfur is lost or transformed into aromatic sulfur at higher ranks (Maes and others, 1997; Gorbaty and Kelemen, 2001).

Verification of Equations to Predict Coal Hydrogen

The geographically specific equations used to predict coal hydrogen are described in table 2. These equations were applied to the PSU-DOE data to verify their accuracy. Figure 6 shows the near 1:1 correspondence between the measured PSU-DOE hydrogen values and the predicted PSU-DOE hydrogen values. Error bars on the figure correspond to an assay reproducibility of 0.3% hydrogen (ASTM, 2000b) and show that most of the scatter can be attributed to the limited precision of the hydrogen assay. The departure of two, low-hydrogen coals (anthracite rank) from the forced regression line suggests that the regression model is not well suited to predict the hydrogen content of anthracite.

Table 2. List of variables, coefficients, and statistics for geographically specific regression equations used to predict the hydrogen content of coal (see equations 2 to 5 in text for variable descriptions).

Data Group	variable name	coefficient		t-statistic	equation statistics
Northern Appalachian	Intercept	-56.22		-14.9	adjusted R^2 = 0.75
	Btu_{dmmf}^2	-2.82	E-07	-16.4	
	Btu_{dmmf}	8.35	E-03	16.4	standard error = 0.18
	$MM_{Parr,dry}$	-5.34	E-02	-49.1	observations = 1028
	lbs S/million Btu	5.97	E-02	12.0	
Central Appalachian	Intercept	-55.81		-18.1	adjusted R^2 = 0.74
	Btu_{dmmf}^2	-2.76	E-07	-19.5	
	Btu_{dmmf}	8.22	E-03	19.7	std. error = 0.19
	$MM_{Parr,dry}$	-5.10	E-02	-39.6	observations = 756
	lbs S/million Btu	1.06	E-01	12.7	
Southern Appalachian	Intercept	-65.88		-13.3	adjusted R^2 = 0.71
	Btu_{dmmf}^2	-3.19	E-07	-14.3	
	Btu_{dmmf}	9.55	E-03	14.4	std. error = 0.21
	$MM_{Parr,dry}$	-5.145	E-02	-36.0	observations = 647
	lbs S/million Btu	7.323	E-02	9.4	
Eastern Interior	Intercept	-41.39		-2.8	adjusted R^2 = 0.73
	Btu_{dmmf}^2	-2.11	E-07	-3.0	
	Btu_{dmmf}	6.30	E-03	3.1	std. error = 0.15
	$MM_{Parr,dry}$	-5.33	E-02	-17.9	observations = 220
	lbs S/million Btu	2.55	E-02	2.9	
Western Interior	Intercept	-4.54		-0.6	adjusted R^2 = 0.82
	Btu_{dmmf}^2	-3.54	E-08	-0.9	
	Btu_{dmmf}	1.21	E-03	1.1	std. error = 0.19
	$MM_{Parr,dry}$	-5.00	E-02	-14.2	observations = 170
	lbs S/million Btu	2.94	E-03	0.3	
Gulf Coast	Intercept	20.97		2.5	adjusted R^2 = 0.73
	Btu_{dmmf}^2	1.35	E-07	2.4	
	Btu_{dmmf}	-2.95	E-03	-2.2	std. error = 0.23
	$MM_{Parr,dry}$	-3.95	E-02	-10.4	observations = 66
	lbs S/million Btu	-5.27	E-02	-1.9	
Rocky Mountain	Intercept	-5.87		-5.4	adjusted R^2 = 0.83
	Btu_{dmmf}^2	-3.39	E-08	-5.7	
	Btu_{dmmf}	1.29	E-03	8.0	std. error = 0.20
	$MM_{Parr,dry}$	-4.31	E-02	-40.5	observations = 641
	lbs S/million Btu	2.41	E-02	1.7	
Northern Great Plains, Pacific Coast	Intercept	1.88		0.5	adjusted R^2 = 0.72
	Btu_{dmmf}^2	5.98	E-09	0.3	
	Btu_{dmmf}	1.64	E-04	0.3	std. error = 0.19
	$MM_{Parr,dry}$	-3.88	E-02	-20.4	observations = 502
	lbs S/million Btu	3.80	E-03	0.3	
High Rank (mvb to lvb)	Intercept	-35.66		5.9	adjusted R^2 = 0.52
	Btu_{dmmf}^2	-1.66	E-07	6.1	
	Btu_{dmmf}	5.20	E-03	-6.0	std. error = 0.28
	$MM_{Parr,dry}$	-4.19	E-02	1.4	observations = 362
	lbs S/million Btu	3.91	E-02	-1.6	
(anthracite)	Intercept	209.20		5.9	adjusted R^2 = 0.67
	Btu_{dmmf}^2	1.02	E-06	6.1	
	Btu_{dmmf}	-2.92	E-02	-6.0	std. error = 0.38
	$MM_{Parr,dry}$	1.74	E-02	1.4	observations = 25
	lbs S/million Btu	-4.78	E-01	-1.6	
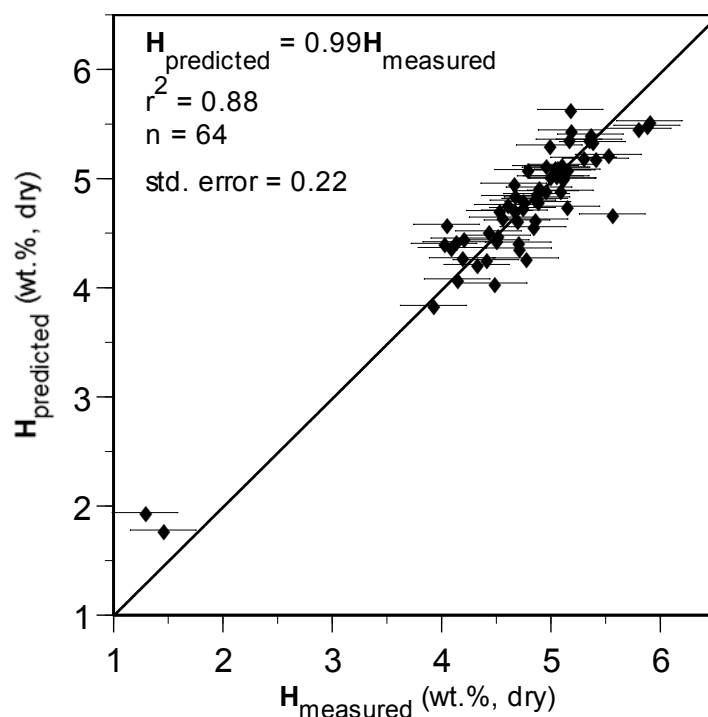
Figure 6. A near 1:1 relationship is observed between the measured PSU-DOE hydrogen values ($H_{measured}$) and predicted PSU-DOE hydrogen values ($H_{predicted}$). The predicted hydrogen values were calculated using equations described in table 2 (in text). The points represent individual PSU-DOE data records selected to have Mott-Spooner difference values within ±250 Btu. Error bars illustrate an assay reproducibility of ±0.3% hydrogen (ASTM, 2000b) and show that most of the scatter is explained by the precision of the hydrogen assay.

Verification of Calculated ICR Net Heating Values

The predicted hydrogen, estimated moisture, and measured Btu values were used with equation 1 to calculate the average net heating value for 169 counties represented in the ICR data set. The county-average results show that the net heating value is about 4.5% less than the gross heating value. This is similar to the 5% difference assumed by the reference method to verify greenhouse gas emissions for the Kyoto Protocol (Houghton and others, 1997). However, as shown in figure 7, the difference between the net and gross heating value varies with coal rank.

The net heating value of lignite is about 10 percent less than its gross heating value; the difference smoothly declines through the coalification series to reach a minimum (1 to 2 percent difference) at the anthracite stage. Figure 7 also shows that the net heating values predicted for the county-average ICR data mimic those calculated using the (measured) PSU-DOE moisture and hydrogen values.
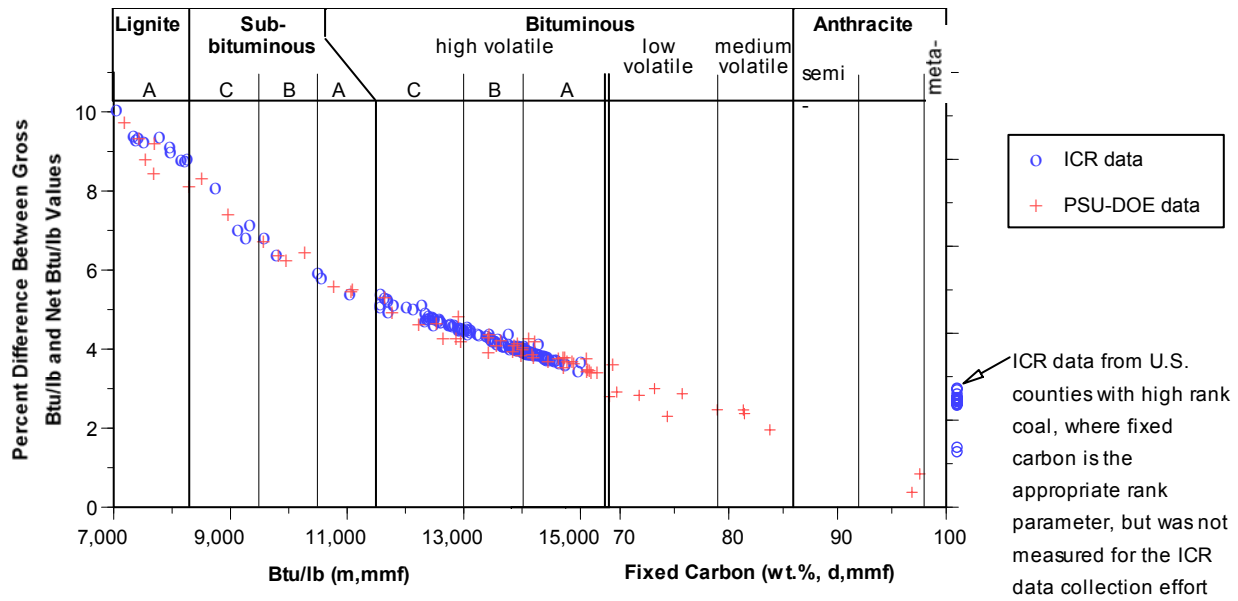
Figure 7. The difference between the net and gross heating value of U.S. coal from two data sets systematically varies with ASTM (1990) coal rank. The percent difference between the gross heating value of coal (Btu_{gross}), and the calculated net heating value (Btu_{net}) corresponds to:

$$Percent\ Difference = \frac{100\ Btu_{gross}}{(Btu_{gross} - Btu_{net})}$$. The PSU-DOE data points represent single coal assays on an equilibrium moisture basis. The ICR data points represent county-average values on an estimated, as-shipped moisture basis.

Parameters for Mapping and Draft Maps (Tasks 5 and 6)

Examination data from fuel and flue gas assays for about 80 electric utility boilers (USEPA, 2003) has shown that mercury capture from flue gas varies according to coal composition, boiler operation and type, and the configuration of pollution emission controls (Pavlish and others, 2003). Various research groups (Chu and others, 2000; Robertson, 2002; ENSR 2003; SAIC, 2003; Maxwell, 2003; AEMS, 2004) have used these assay data to develop regression equations that predict the efficiency of mercury removal from flue gas for power plants classified by boiler type and emission controls. Independent variables used in these equations include the coal heating value, chlorine content, and sulfur content.

Figures 8, 9, and 10 are maps that respectively show the potential uncontrolled sulfur, mercury, and chlorine emissions of U.S. coal by county-of-origin. These potential emission rates were calculated using the ICR data and equations for parameters A, B, and C listed table 3. Similar maps (figures 11 to 15) show predicted mercury emissions for coal burned in power plants classified by emission control technology; these maps were created using the ICR data and equations from SAIC (2003), which correspond to parameters D, E, F, G, and H listed in table 3. Finally, maps showing output-based mercury emissions (lbs Hg/gigawatt-hour) for U.S. coal by county-of-origin were made for two emission control technologies (cold-side ESP-FGD technology, and fabric filter technology; figures 16 and 17). The output based emission rates were calculated using ICR data expressed on a net energy basis, SAIC equations, and a nominal heat rate of 35% (parameters I and J, table 3).

Table 3. Selected parameters for mapping.

	Parameter	Emission Control	Parameter Calculation
input-based calculation	**A** lbs S/10^6 Btu	uncontrolled	$= 10^6/\text{Btu} \times \text{S}/100$
	B lbs Hg/10^{12} Btu	uncontrolled	$= 10^{12}/\text{Btu} \times \text{Hg}/10^6$ (note: $\mathbf{B_{net}}$ is calculated using Btu_{net} and Hg_{moist})
	C lbs Cl/ 10^9 Btu	uncontrolled	$= 10^9/\text{Btu} \times \text{Cl}/10^6$
	D lbs Hg/10^{12} Btu	cold-side ESP	$= \mathbf{B} \times [1 - \exp(0.031 - 0.003929 \times \mathbf{C/A})]$
	E lbs Hg/10^{12} Btu	cold-side ESP + FGD	$= \mathbf{B} \times [1 - \exp(1.8529 - 0.27149 \times \ln(\mathbf{C} \times 1000))]$
	F lbs Hg/10^{12} Btu	hot-side ESP	$= \mathbf{B} \times [1 - \exp(0.0759 - 0.003816 \times \mathbf{C})]$
	G lbs Hg/10^{12} Btu	hot-side ESP + FGD	$= \mathbf{B} \times [1 - \exp(2.7019 - 0.29952 \times \ln(\mathbf{C} \times 1000))]$
	H lbs Hg/10^{12} Btu	SDA + FF	$= \mathbf{B} \times [1 - \exp(10.7111 - 1.22628 \times \ln(\mathbf{C} \times 1000))]$
output-based calculation	**I** lbs Hg/TW-h	cold-side ESP + FGD	$= \mathbf{B_{net}} \times [1 - \exp(1.8529 - 0.27149 \times \ln(\mathbf{C} - 1000))] \times 10.26$
	J lbs Hg/TW-h	SDA + FF	$= \mathbf{B_{net}} \times [1 - \exp(10.7111 - 1.22628 \times \ln(\mathbf{C} \times 1000))] \times 10.26$

Notes to Table:

ESP   Electrostatic Precipitator

FGD   wet Flue Gas Desulfurization

SDA   Spray Dry Adsorption

FF   Fabric Filter

Btu   gross British thermal units per pound (ICR data, dry basis)

S   wt.% Sulfur (ICR data, dry basis)

Hg   µg/g Mercury (ICR data, dry basis)

Cl   µg/g Chlorine (ICR data, dry basis)

$\mathbf{B_{net}}$ pounds Hg/10^{12} Btu$_{net}$

Hg_{moist} is the ICR mercury value (dry µg/g Hg) adjusted to a moist basis according to:
Hg_{moist} = Hg (100-M)/100, using estimated moisture (M) values for the ICR data aggregated by county-of-origin.

Btu_{net} is the ICR gross heating value (dry, Btu/lb) adjusted to a net, moist basis according to: Btu_{net} = (Btu $\times$ (100-M)/100) – 90.3 $\times$ [(M $\times$ 0.1119)+H], where M is the estimated moisture for the ICR data aggregated by county-of-origin, H is the corresponding moist-basis hydrogen value (also estimated) which excludes hydrogen in moisture.

Equations for parameters **D** to **J** are adopted from table 4-3 in SAIC (2003), with the coefficient used for parameters **I** and **J** (10.26) selected to approximate a heat rate of 35% (exactly 9,750 gross Btu/kilowatt-hour, which is approximately 10,260 net Btu/kilowatt-hour).

KEY

Coalfield extent

Sulfur Emissions, uncontrolled potential, lbs S per million Btu

0.30 - 0.40
0.40 - 0.60
0.60 - 0.83
0.83 - 1.67
1.67 - 2.50
2.50 - 3.75
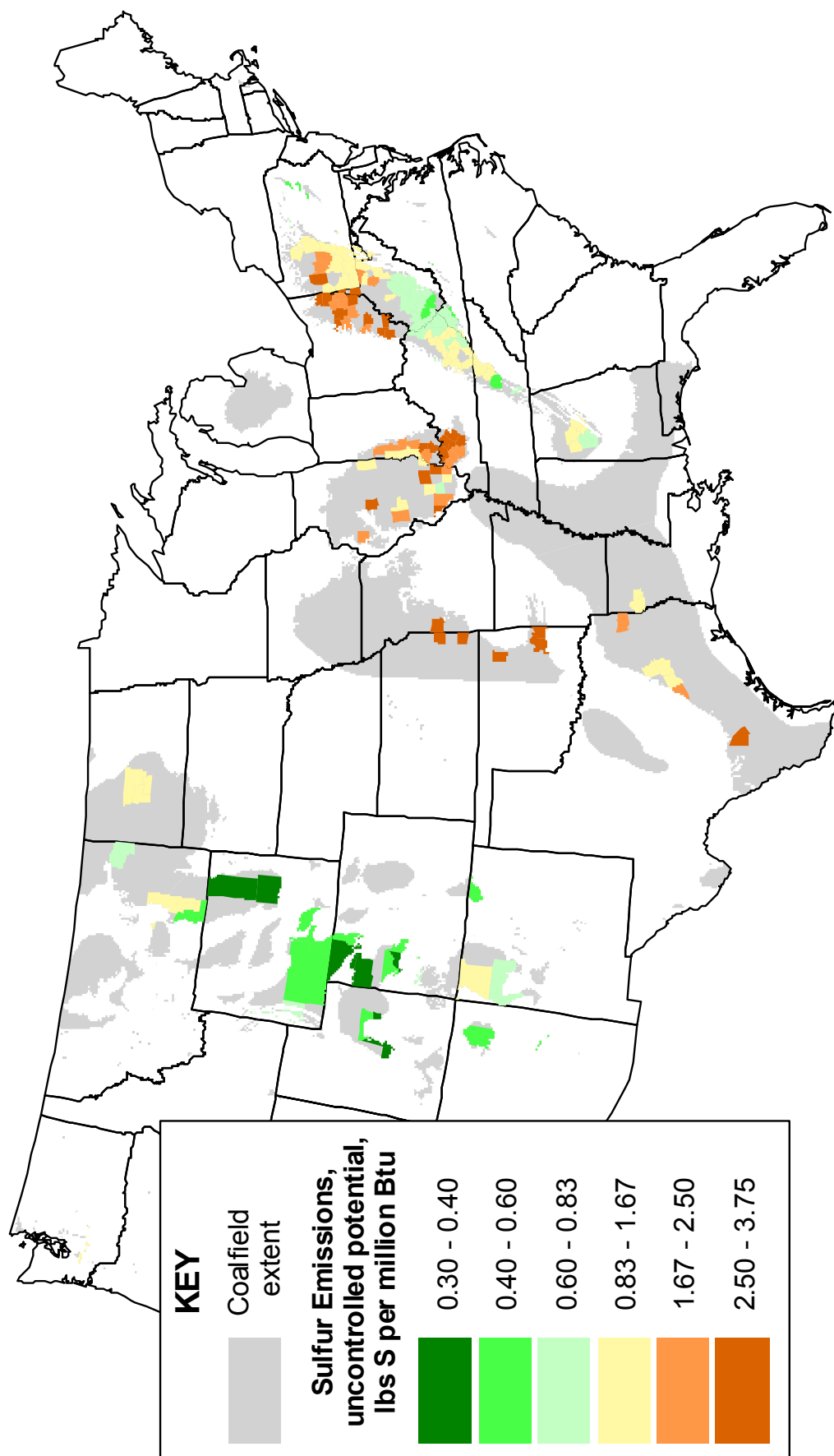
Figure 8. Potential uncontrolled sulfur emissions from coal (lbs S/10^6 Btu) by U.S. county-of-origin (county-average values calculated using selected ICR data).

KEY

Coalfield extent

Mercury Emissions, uncontrolled potential, lbs Hg per trillion Btu

< 2

2 - 4

4 - 5.8

5.8 - 9.2

9.2 - 15

15 - 30

30 - 52
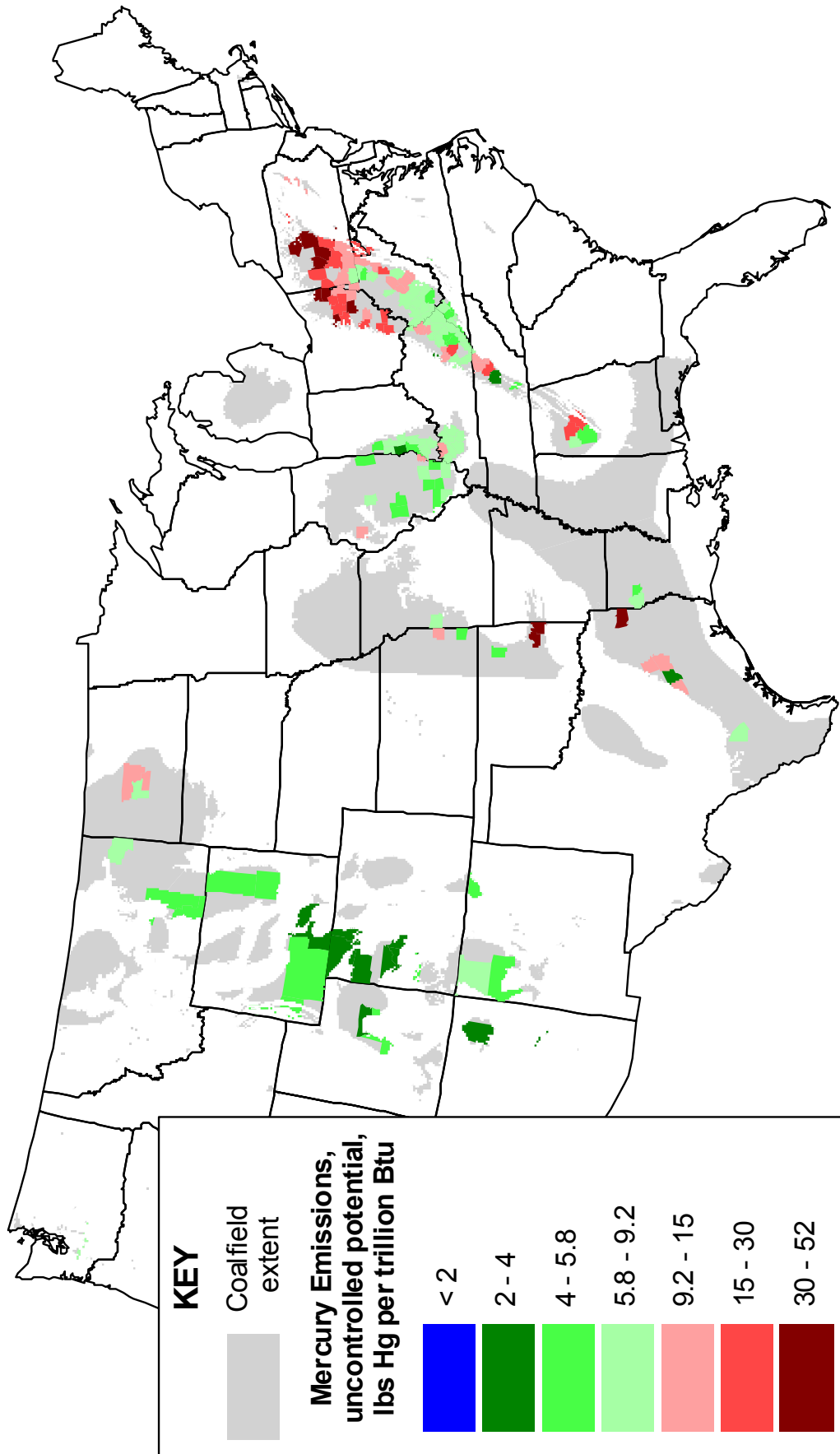
Figure 9. Potential uncontrolled mercury emissions from coal (lbs Hg/10^{12} Btu) by U.S. county-of-origin (county-average values calculated using selected ICR data).
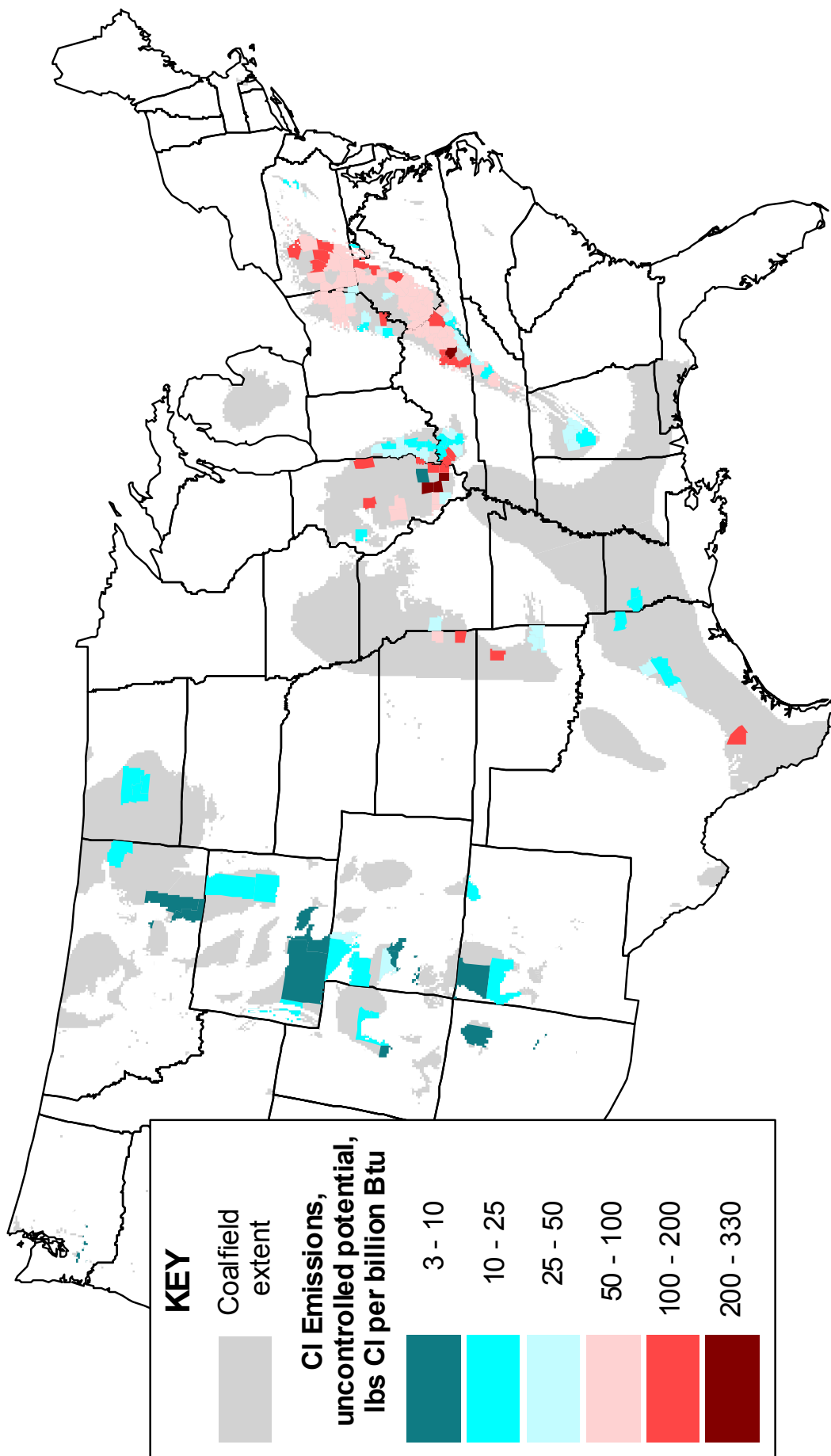
Figure 10. Potential uncontrolled chlorine emissions from coal (lbs Cl/10^9 Btu) by U.S. county-of-origin (county-average values calculated using selected ICR data).

KEY

Coalfield extent

Cl Emissions, uncontrolled potential, lbs Cl per billion Btu

3 - 10
10 - 25
25 - 50
50 - 100
100 - 200
200 - 330
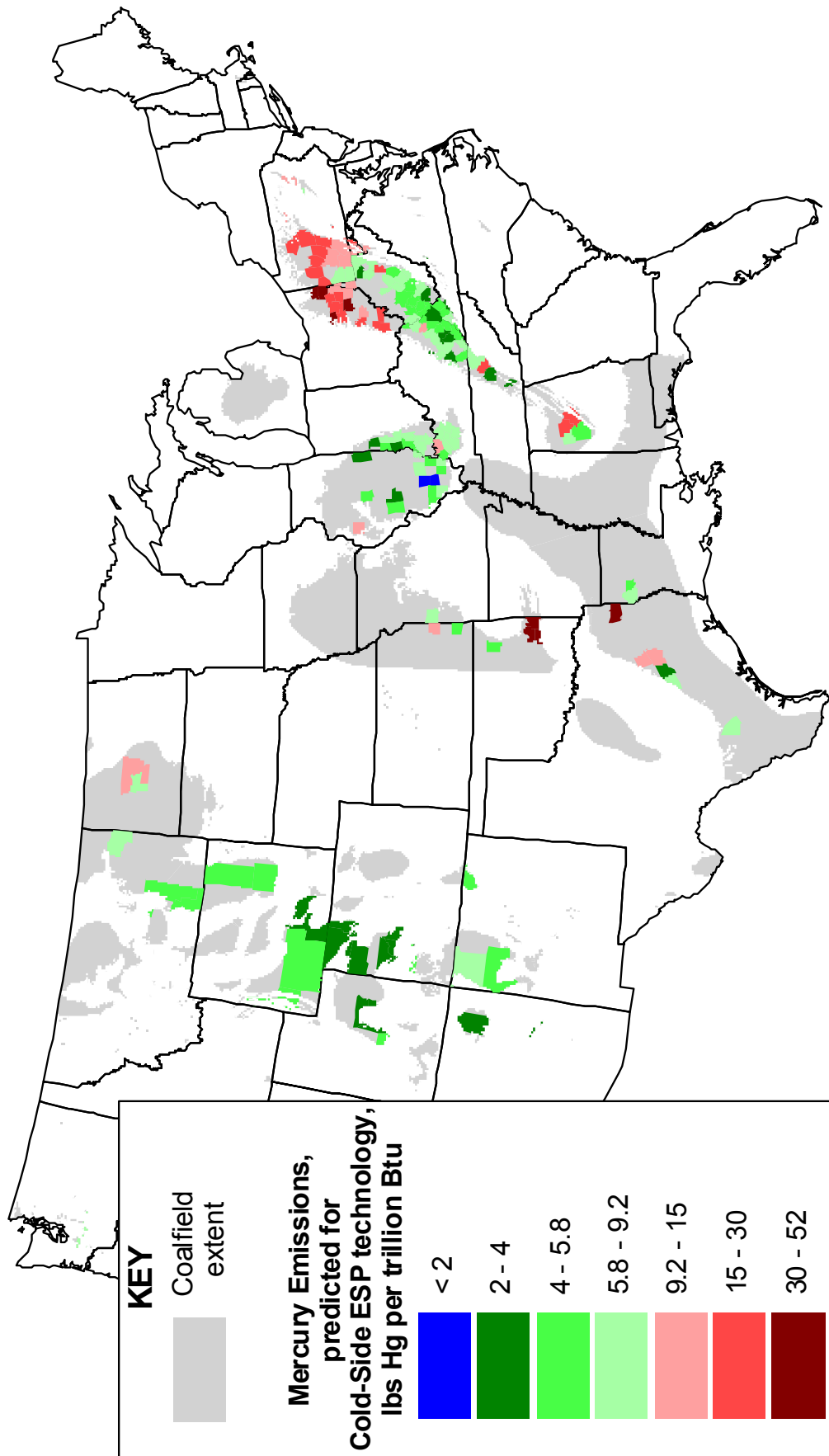
Figure 11. Predicted mercury emissions (lbs Hg/10^{12} Btu) for coal burned in electric utilities with Cold-Side, Electrostatic Precipitators (Cold-Side ESP technology). The map shows county-average values, which were calculated using selected ICR data, and an equation from SAIC (2003) listed as parameter D, table 3, this report.

KEY

Coalfield extent

Mercury Emissions, predicted for Cold-Side ESP technology, lbs Hg per trillion Btu

< 2
2 - 4
4 - 5.8
5.8 - 9.2
9.2 - 15
15 - 30
30 - 52

KEY

Coalfield extent

Mercury Emissions, predicted for Cold-Side ESP-FGD technology, lbs Hg per trillion Btu

< 2
2 - 4
4 - 5.8
5.8 - 9.2
9.2 - 15
15 - 30
30 - 52

Figure 12. Predicted mercury emissions (lbs Hg/10^{12} Btu) for coal burned in electric utilities with Cold-Side, Electrostatic Precipitators and Flue Gas Desulfurization controls (Cold-Side ESP-FGD technology). The map shows county-average values, which were calculated using selected ICR data, and an equation from SAIC (2003) listed as parameter E, table 3, this report.

Figure 13. Predicted mercury emissions (lbs Hg/10^{12} Btu) for coal burned in electric utilities with Hot-Side, Electrostatic Precipitators (Hot-Side ESP technology). The map shows county-average values, which were calculated using selected ICR data, and an equation from SAIC (2003) listed as parameter F, table 3, this report.

KEY

Coalfield extent

Mercury Emissions, predicted for Hot-Side ESP technology, lbs Hg per trillion Btu

< 2
2 - 4
4 - 5.8
5.8 - 9.2
9.2 - 15
15 - 30
30 - 52

KEY

Coalfield extent

Mercury Emissions, predicted for Hot-Side ESP - FGD technology, lbs Hg per trillion Btu

< 2

2 - 4

4 - 5.8

5.8 - 9.2

9.2 - 15

15 - 30

30 - 52

Figure 14. Predicted mercury emissions (lbs Hg/10^{12} Btu) by U.S. county-of-origin for coal burned in electric utilities with Hot-Side, Electrostatic Precipitators and Flue Gas Desulfurization controls (Hot-Side ESP-FGD technology). The map shows county-average values which were calculated using selected ICR data, and an equation from SAIC (2003) listed as parameter G, table 3, this report.
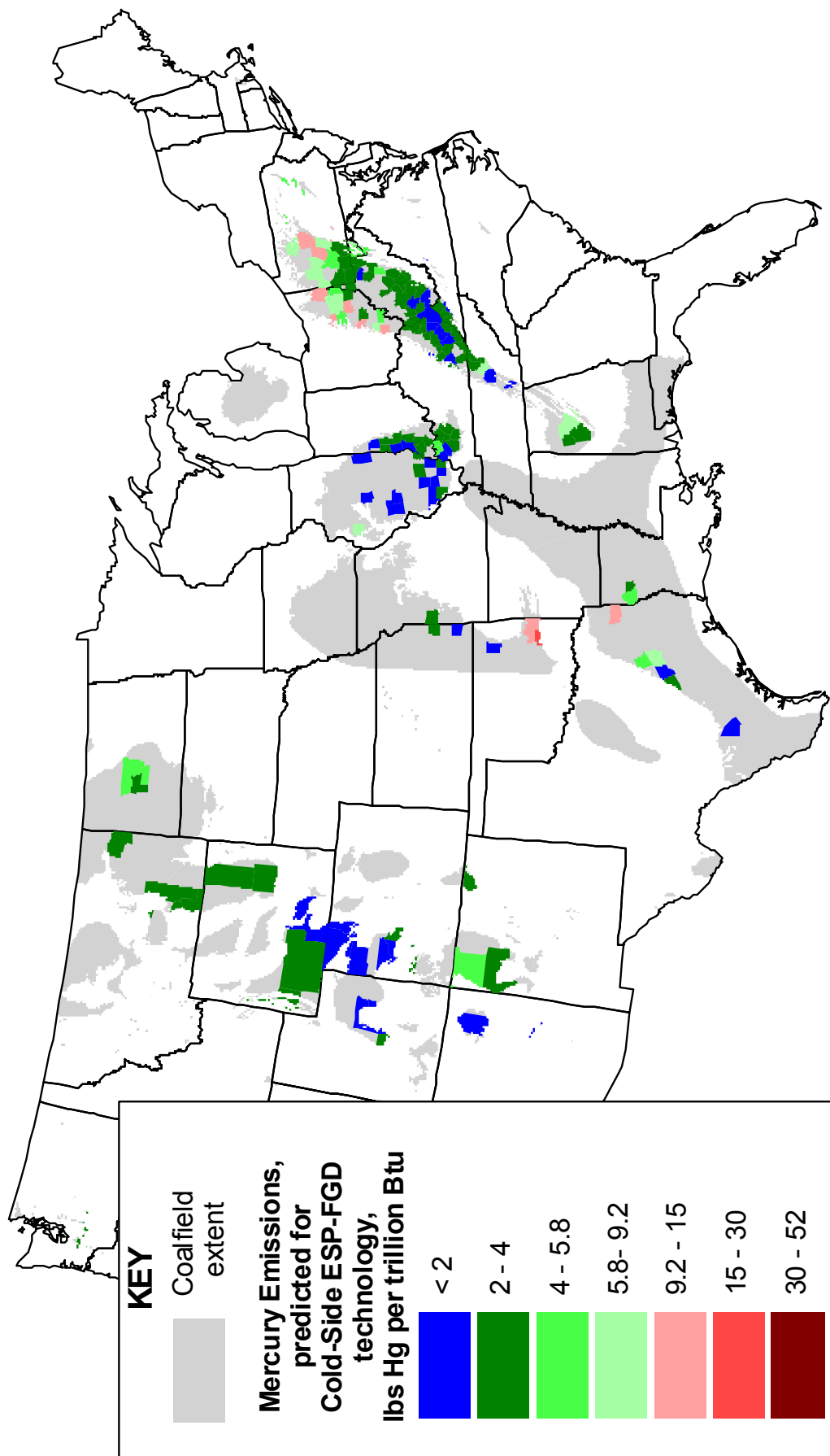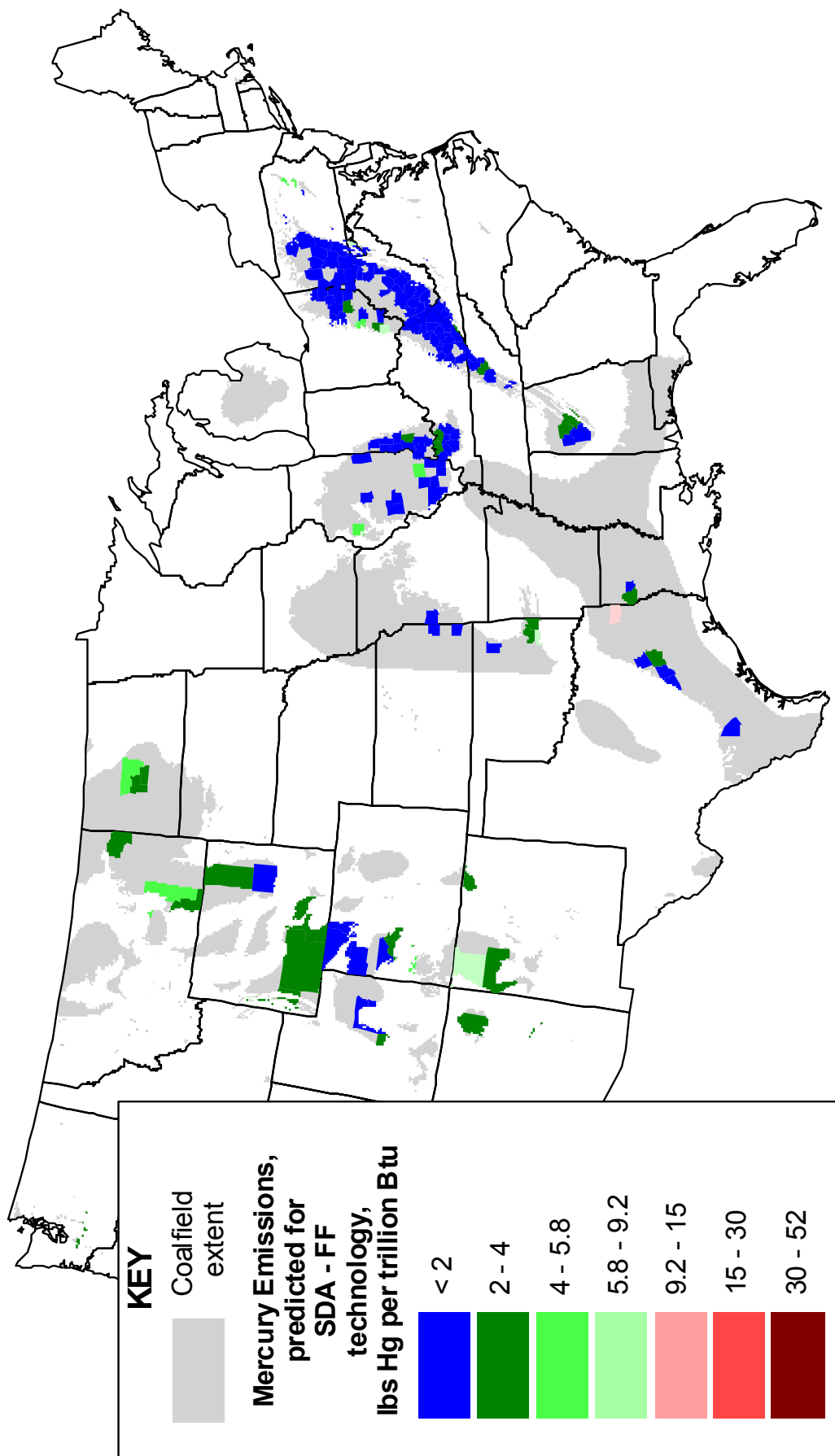
Figure 15. Predicted mercury emissions (lbs Hg/10^{12} Btu) by U.S. county-of-origin for coal burned in electric utilities with Spray-Dry Adsorption and Fabric Filter controls (SDA-FF technology). The map shows county-average values, which were calculated using selected ICR data, and an equation from SAIC (2003) listed as parameter H, table 3, this report.
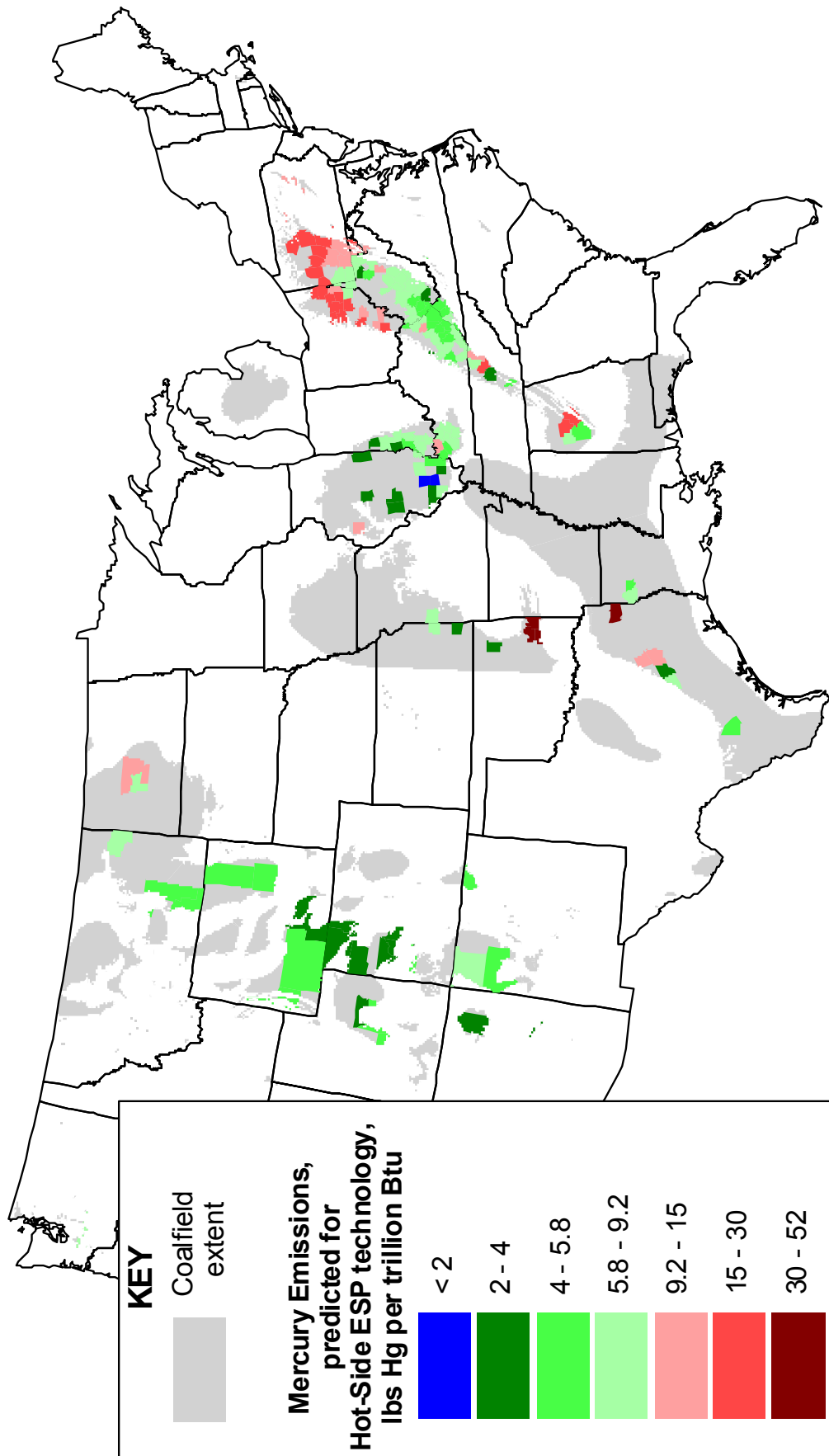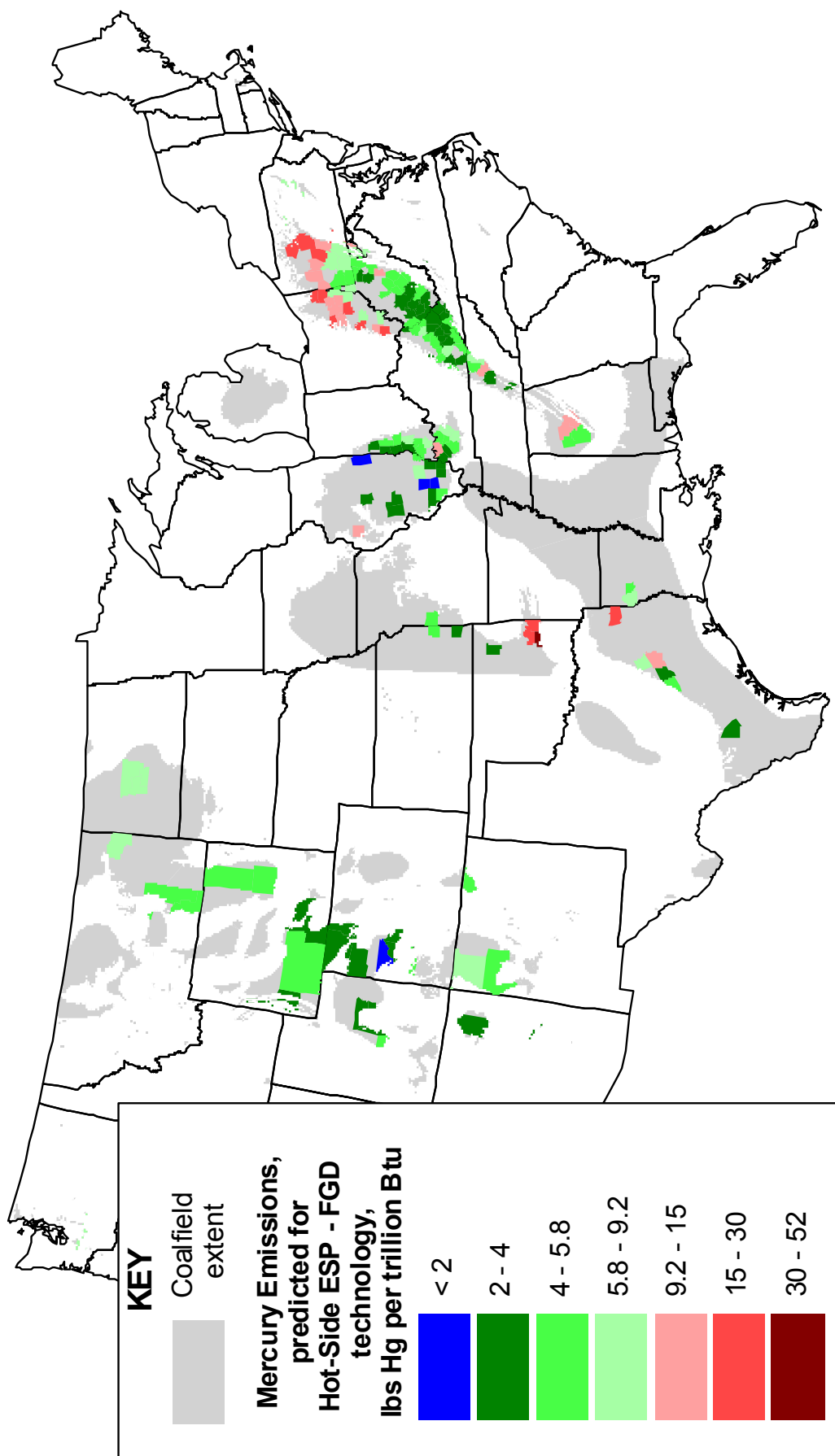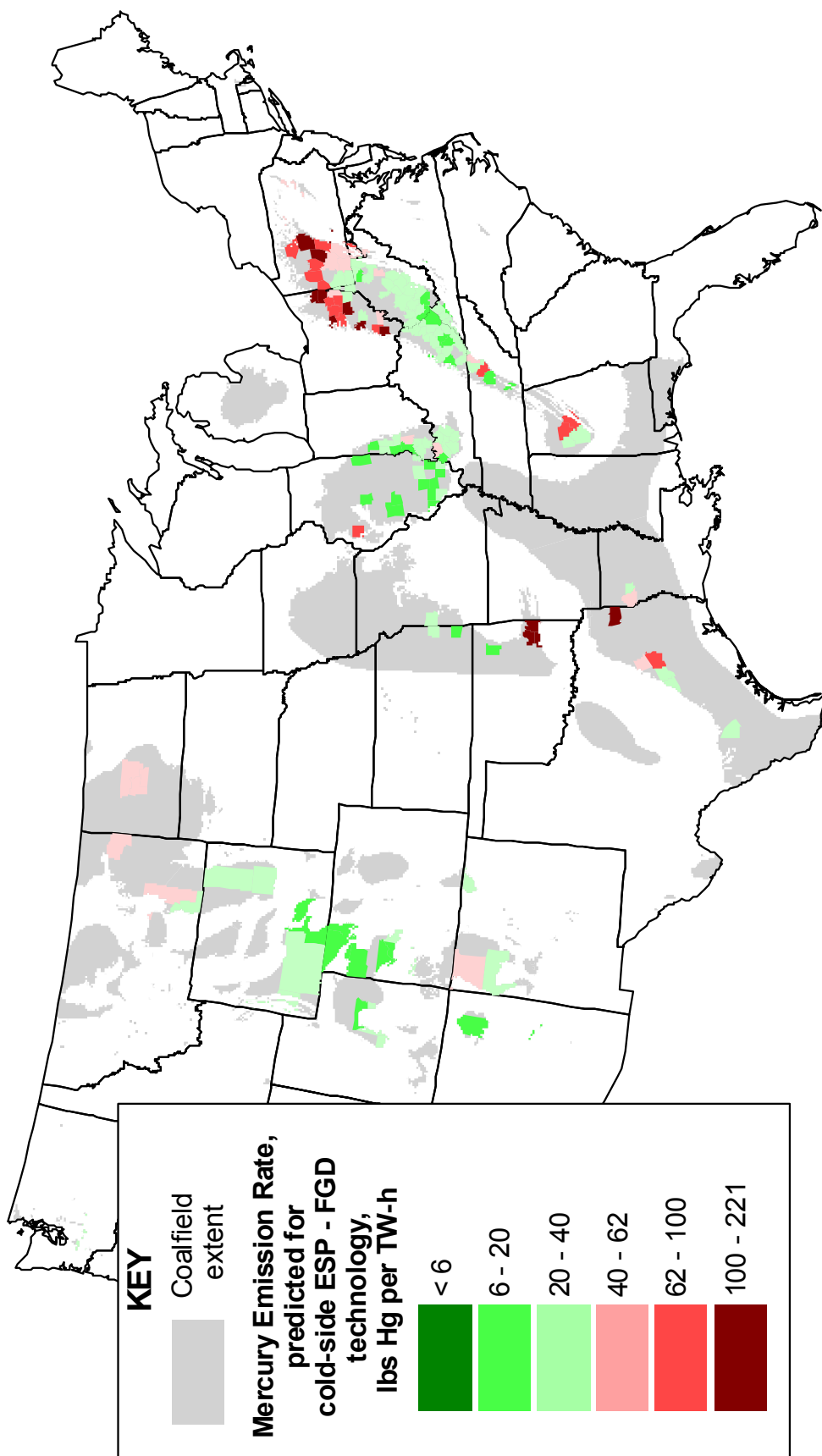
Figure 16. Predicted mercury emission rate (lbs Hg/terawatt-hour, which is the same as lbs Hg x 10^{-6}/megawatt-hour) for coal burned in electric utilities with Cold-Side, Electrostatic Precipitators and wet Flue Gas Desulfurization controls (Cold-Side ESP-FGD technology). The map shows county-average values, which were calculated using selected ICR data expressed on a net energy basis, a nominal heat rate of 35%, and an equation from SAIC (2003) listed as parameter l, table 3, this report.

KEY

Coalfield extent

Mercury Emission Rate, predicted for cold-side ESP - FGD technology, lbs Hg per TW-h

< 6
6 - 20
20 - 40
40 - 62
62 - 100
100 - 221

Figure 17. Predicted mercury emission rate (lbs Hg/terawatt-hour, which is the same as lbs Hg x 10^{-6}/megawatt-hour) for coal burned in electric utilities with Spray-Dry Adsorption and Fabric Filter controls (SDA-FF technology). The map shows county-average values, which were calculated using selected ICR data expressed on a net energy basis, a nominal heat rate of 35%, and an equation from SAIC (2003) listed as parameter J, table 3, this report.

KEY

Coalfield extent

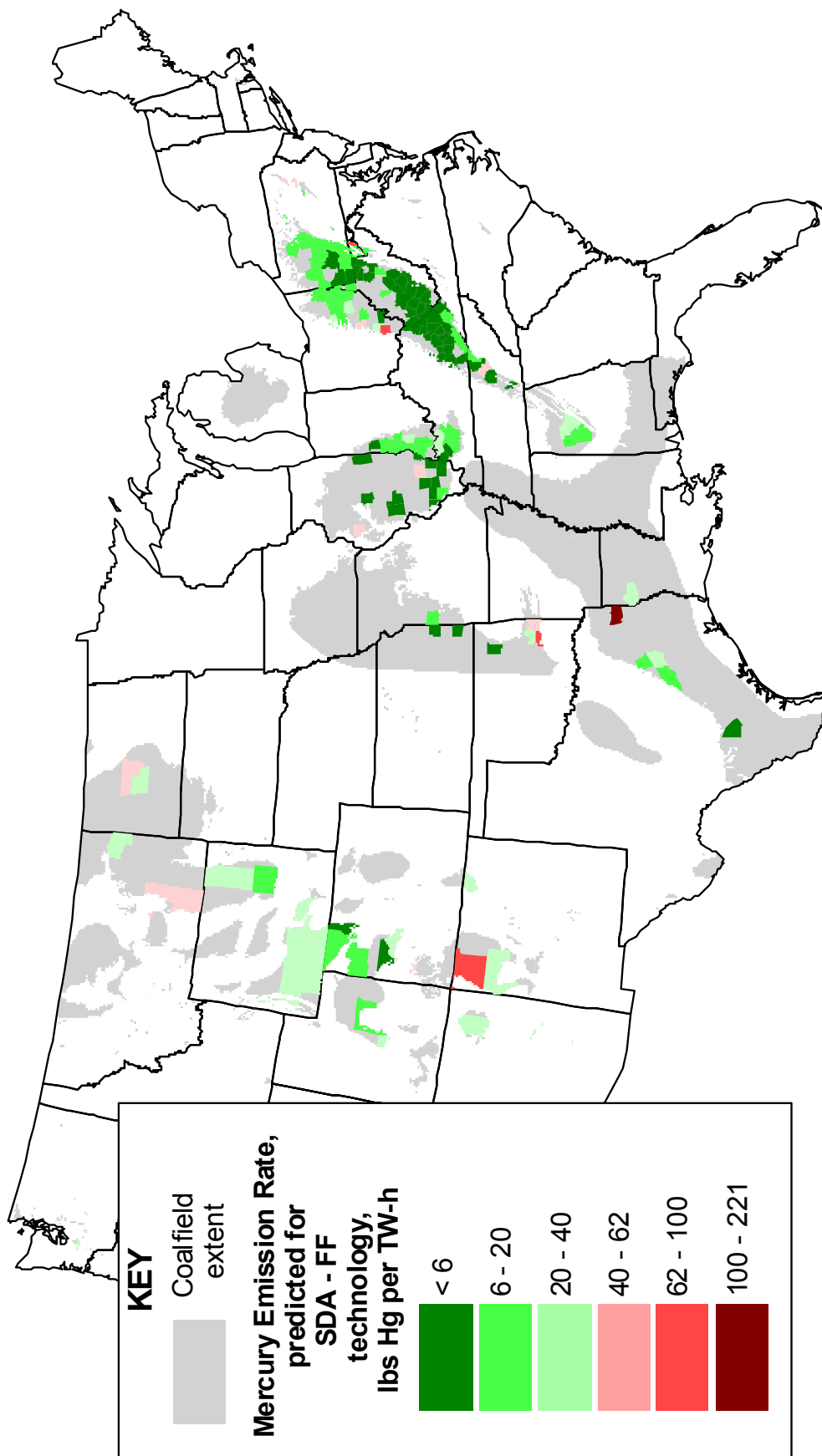**Mercury Emission Rate, predicted for SDA - FF technology, lbs Hg per TW-h**

< 6

6 - 20

20 - 40

40 - 62

62 - 100

100 - 221

RESULTS AND DISCUSSION

Draft maps showing the potential mercury and acid-gas emissions from coal combustion, by U.S. county-of-origin were constructed using selected ICR coal quality data, and technology-specific equations that predict mercury capture (SAIC, 2003). In this section we evaluate the coal assay data that these maps are based on and conclude with a brief discussion of the significance and limitations of the draft maps.

Evaluation of Coal Assay Data

Coal assay data used in this study include:

- 19,507 FERC 423 data records from 187 U.S. counties (USEIA, 2003a),

- 25,818 ICR data records from 169 U.S. counties (USEPA, 2003),

- 5,602 CTRDB data records from 116 U.S. counties (USEIA, 2003b),

- 5,045 COALQUAL data records from 340 U.S. counties (Bragg and others, 1997), and

- 73 PSU-DOE data records from 47 U.S. counties (Anonymous, 1990; Davis and Glick, 1993; Scaroni and others, 1999).

The ICR data are the foundation of the draft maps (figures 8 to 17), whereas the COALQUAL, FERC 423, CTRDB, and PSU-DOE data were used to estimate ICR moisture and hydrogen values, and to verify these estimates and their derived values. Comparison of these data sets shows data limitations, provides geochemical insights, and suggests mercury mitigation strategies.

County-average, moisture, ash, sulfur, and Btu/lb values for four data sets are compared in figures 18 and 19. Note that the data sets compared in figure 18 are populated by different numbers of counties, whereas the comparisons shown in figure 19 only include counties that are common to both the ICR, and the FERC 423, CTRDB, or COALQUAL data sets.
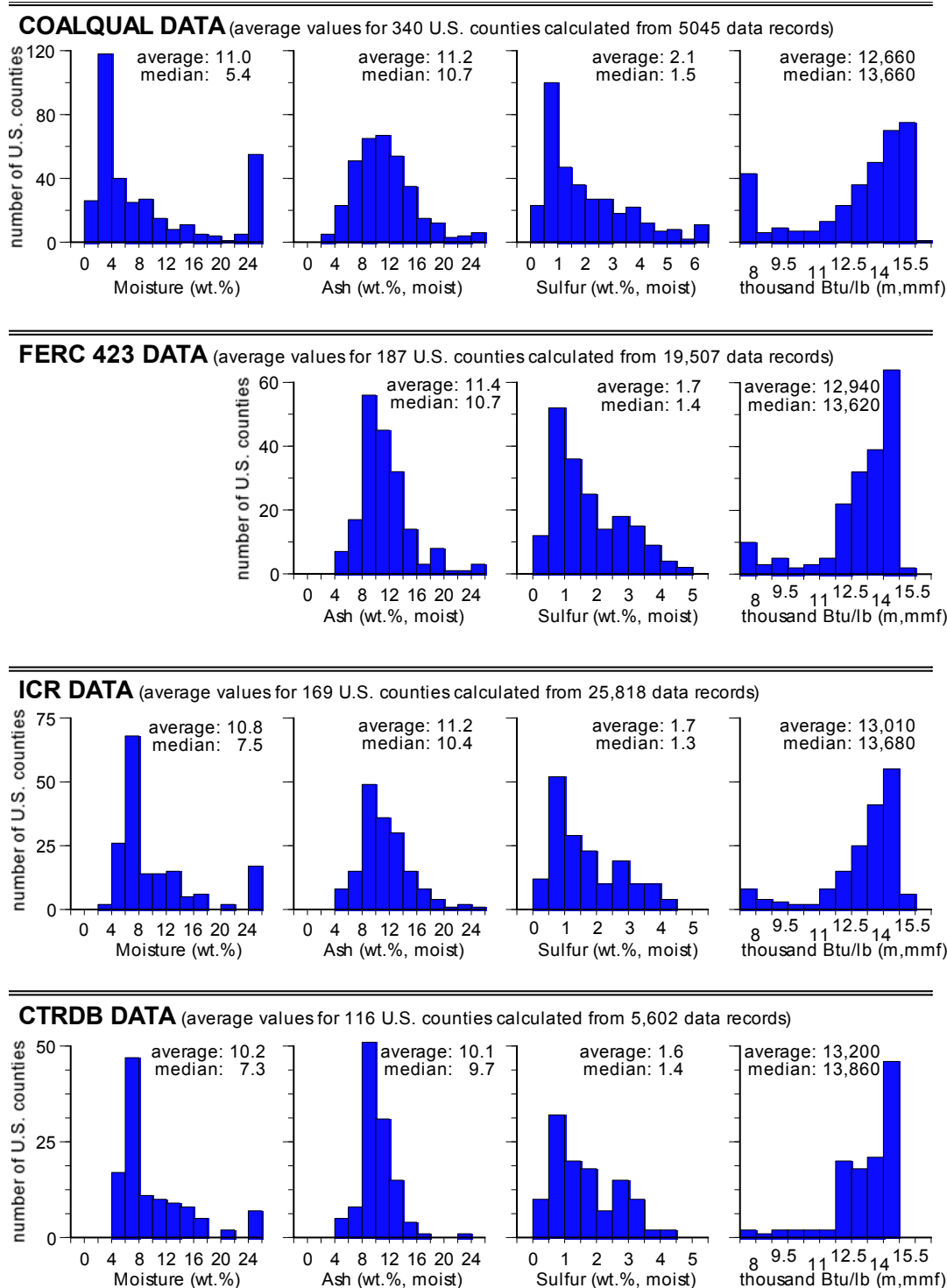
Figure 18. Histograms showing the distribution of county-average coal quality values for the COALQUAL, FERC 423, ICR, and CTRDB data sets. Moisture, ash, and sulfur values are expressed on a moist, whole-coal basis, whereas the Btu/lb values are expressed on a moist, mineral-matter-free basis, calculated after ASTM (1990).

30
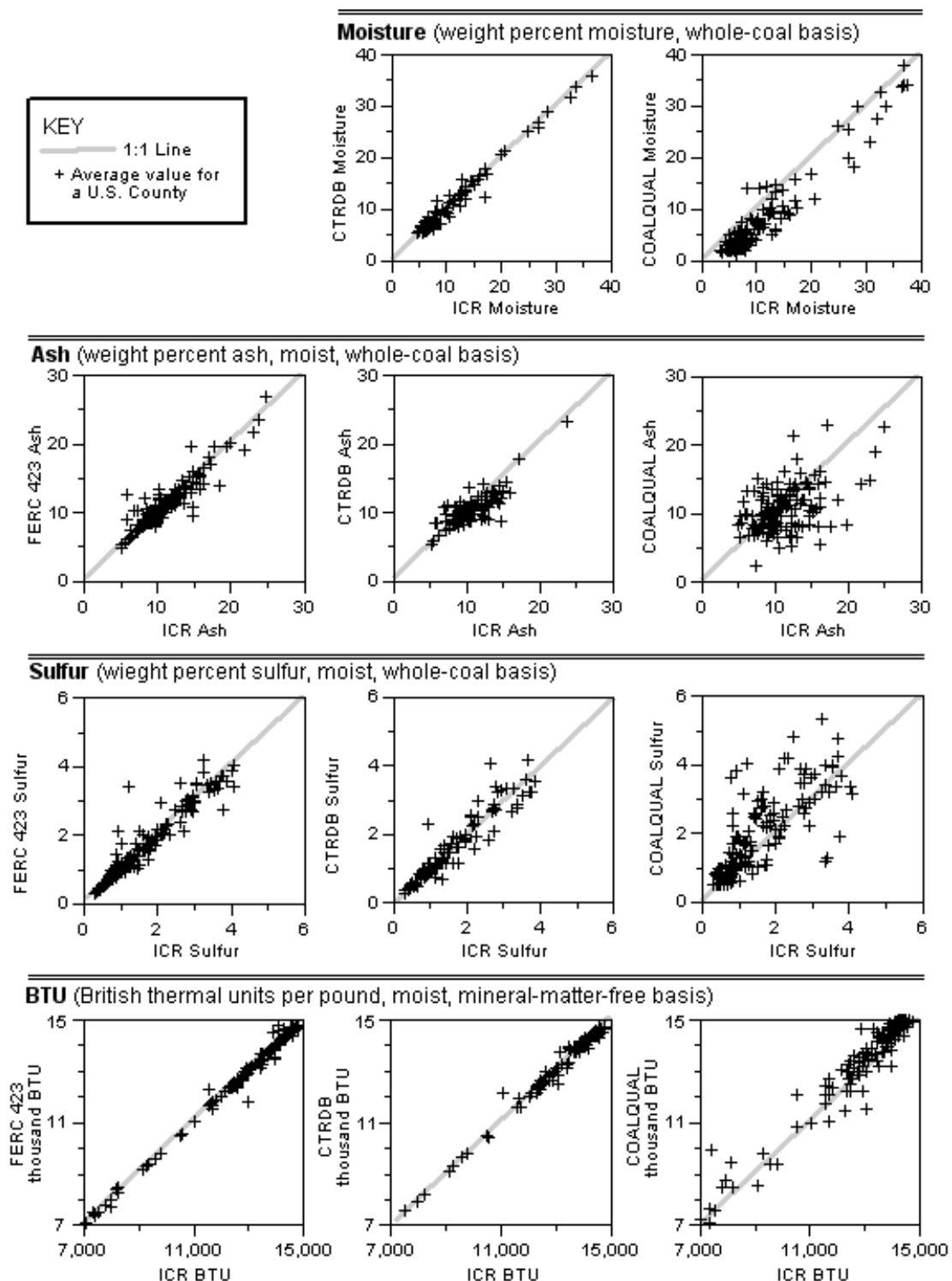
Figure 19. Cross-plots comparing the county-average moisture, ash, sulfur, and Btu values from the ICR data set with those from the CTRDB, COALQUAL, and FERC 423 data sets; the Btu/lb values were calculated after ASTM (1990).

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Figures 18 and 19 show reasonably good agreement between the data sets, especially for data corresponding to commercial coal shipments (FERC 423, CTRDB, and ICR). The correlation between the ICR and FERC 423 sulfur values shown in figure 19 deserves comment. Despite the good correlation, a few counties deviate from the 1:1 line. Many of these deviations can be attributed to instances where the county-average values are calculated from one or two data records. However a few instances may indicate potential bias in ICR data. Given that the ICR data relied on periodic assays, and include a disproportionate number of records for small (<50 MW) utilities, it is likely that the FERC 423 data better represent the quality of commercial U.S. coal than the ICR data. Moreover, sulfur exhibits a positive correlation with mercury for aggregated data (Quick and others, 2003). Consequently, instances where ICR sulfur is higher than FERC 423 sulfur may indicate erroneously high county-average ICR mercury values. Conversely, instances where the ICR sulfur is lower than the FERC 423 sulfur may indicate erroneously low county-average ICR mercury values.

The larger number of counties included in the COALQUAL data set should be considered when evaluating the data distributions shown in figure 18. For example, the relatively high, average moisture value for the 340 counties listed in the COALQUAL data set (figure 18) is a result of the comparatively large number of counties in the COALQUAL data set with high-moisture (low-rank) coal. Thus, the relatively high average COALQUAL moisture value shown in figure 18 is due to a geographic, rather than analytical, bias. Restricting the comparison of moisture values to common counties (figure 19) shows that the COALQUAL assay moisture values are actually relatively low. Although the relatively low COALQUAL moisture values may relate to added moisture from washing of commercial coal (ICR and CTRDB data), moisture loss prior to analysis of the COALQUAL coal samples is probably more

significant. Indeed, Bragg and others (1997) noted that the calculated ASTM rank for some

COALQUAL data records might be anomalously high due to air-drying of the samples before

analysis. The low COALQUAL moisture values due to assay bias are also consistent with the

relatively high, moist-basis COALQUAL Btu/lb values (figure 19).[2]

As noted earlier in this report, systematically low COALQUAL moisture values

complicate the evaluation of rank and the calculation of net heating values. Fortunately, the low

moisture values have little effect on COALQUAL emission factors expressed on an energy basis.

For example, the calculation of pounds sulfur per million Btu gives the same result regardless of

whether moist-basis sulfur and Btu/lb values, or dry-basis sulfur and Btu/lb values, are used for

the calculation. Figures 20 and 21 compare ICR sulfur, mercury, and chlorine values expressed

on an energy-basis to equivalent COALQUAL values.

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[2] As noted in an earlier report (Quick and others, 2004), it was necessary to adjust some COALQUAL assay values (notably Hg and Cl) for unmeasured residual moisture in the analysis specimen. This systematic bias was the result of the sample preparation method for inorganic assays, and is not related to the moisture bias described here (which, with few exceptions, only influences the proximate, ultimate, and sulfur form analyses).
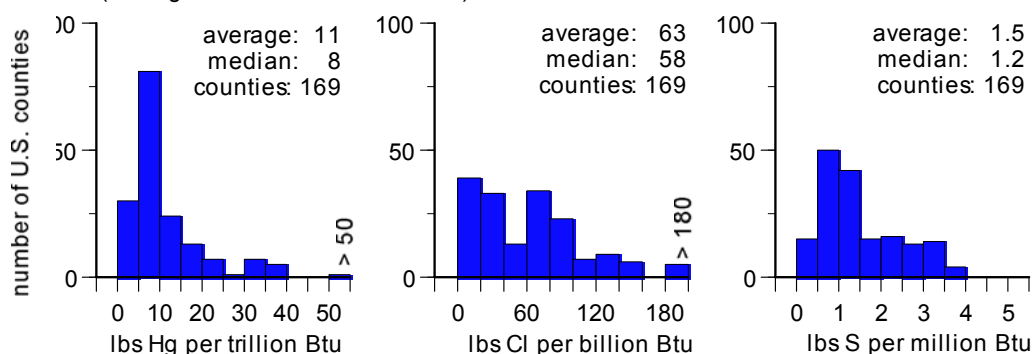
Figure 20. Distribution of county-average, mercury, chlorine and sulfur values for in-ground coal (COALQUAL DATA) and commercial coal (ICR DATA).
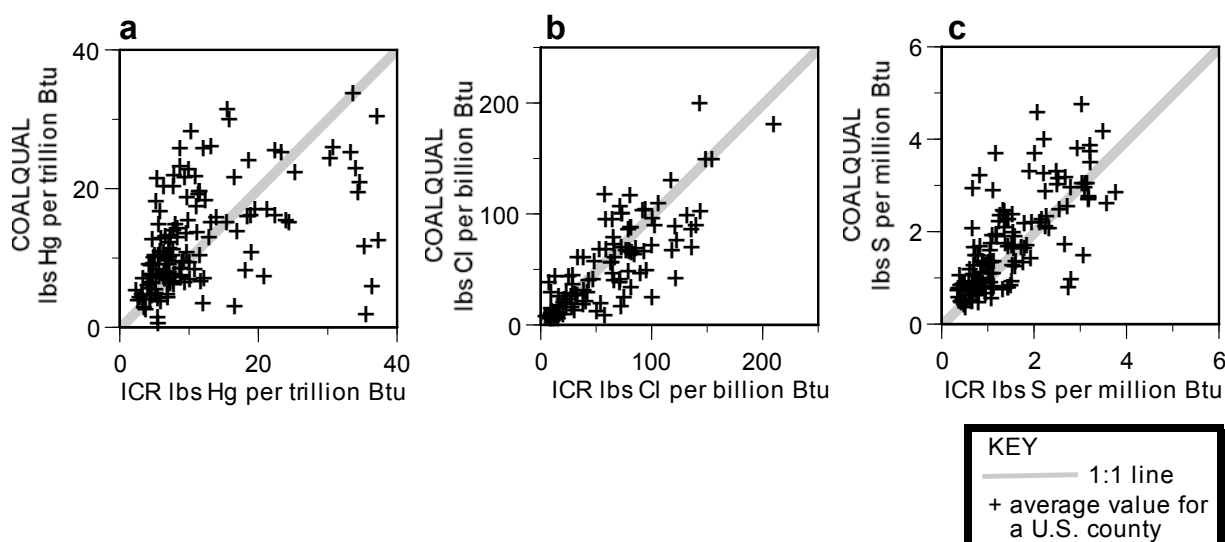

Figure 21. Comparison of mercury, chlorine, and sulfur values in the ICR and COALQUAL data sets. Data points show average values for U.S. counties common to both data sets.

When examining figures 20 and 21 it is useful to recognize that the COALQUAL data indicate the quality of the in-ground coal resource, whereas the ICR data indicate the quality of commercial coal produced during 1999. Differences between the COALQUAL and ICR data are inevitable because the COALQUAL data include additional records for coal beds that are not mined. Nonetheless, comparison of these data is instructive. Figure 20 shows higher sulfur and mercury values for the COALQUAL data than the ICR data. Quick and others (2003) also observed higher COALQUAL sulfur and mercury values, which they attributed to selective mining of low-sulfur and low-mercury coal, as well as reduction of sulfur and mercury due to washing of mined coal. However, figure 21a shows that the mercury content of in-ground coal (COALQUAL data) is not always lower than the mercury content of commercially shipped coal (ICR data) when the comparison is restricted to coal from common counties-of-origin. Counties where the mined coal contains more mercury than indicated by the COALQUAL data are colored red in figure 22. The reason for the higher mercury content of coal mined in these areas may be the combined result of limited washing, and contamination of mined coal by high-mercury partings, roof rock, or floor rock; these contaminants are generally not included in COALQUAL assay specimens because USGS sample collections guidelines (Swanson and Huffman, 1976) require partings more than 5 mm thick to be excluded from the analysis sample. Increased coal washing may be an effective Hg reduction strategy in instances where the ICR mercury values are greater than the COALQUAL mercury values. Green areas on figure 22 show where mined coal contains substantially less Hg than the in-ground resource. Selective mining and/or extensive coal washing probably explain these occurrences. For a few counties, these differences may simply indicate bias in the ICR data (suggested by the different FERC 423

and ICR sulfur values, discussed above) or instances where the county average values are based on only a few data records.

The different chlorine distributions for the COALQUAL and ICR data shown in figure 20 suggest preferential mining of counties with high-chlorine coal. However, such inferences are uncertain given the limitations of the chlorine assays. For example, nearly 30% of the COALQUAL chlorine values are reportedly below the assay detection limit (Bragg and others, 1997). Although only 14% of the selected ICR records are reportedly below the detection limit, this is probably a minimum value. Nyberg (2003) notes that methods used to determine chlorine concentrations in the ICR data collection effort were unreliable below 200 parts-per-million (ppm or $\mu g/g$). Thirty percent of the selected ICR data records show dry chlorine at or below 200 ppm. Moreover, figure 23 shows that western U.S. counties are responsible for a disproportionate share of the low-chlorine ICR data records.

KEY

Coalfield extent

Difference between Hg content of in-ground coal (COALQUAL data) and Hg in mined coal (ICR data) lbs Hg per trillion Btu

MORE Hg in mined coal than in-ground coal

+33 to +15
+15 to +5
+5 to -5
-5 to -10
-10 to -30

LESS Hg in mined coal than in-ground coal
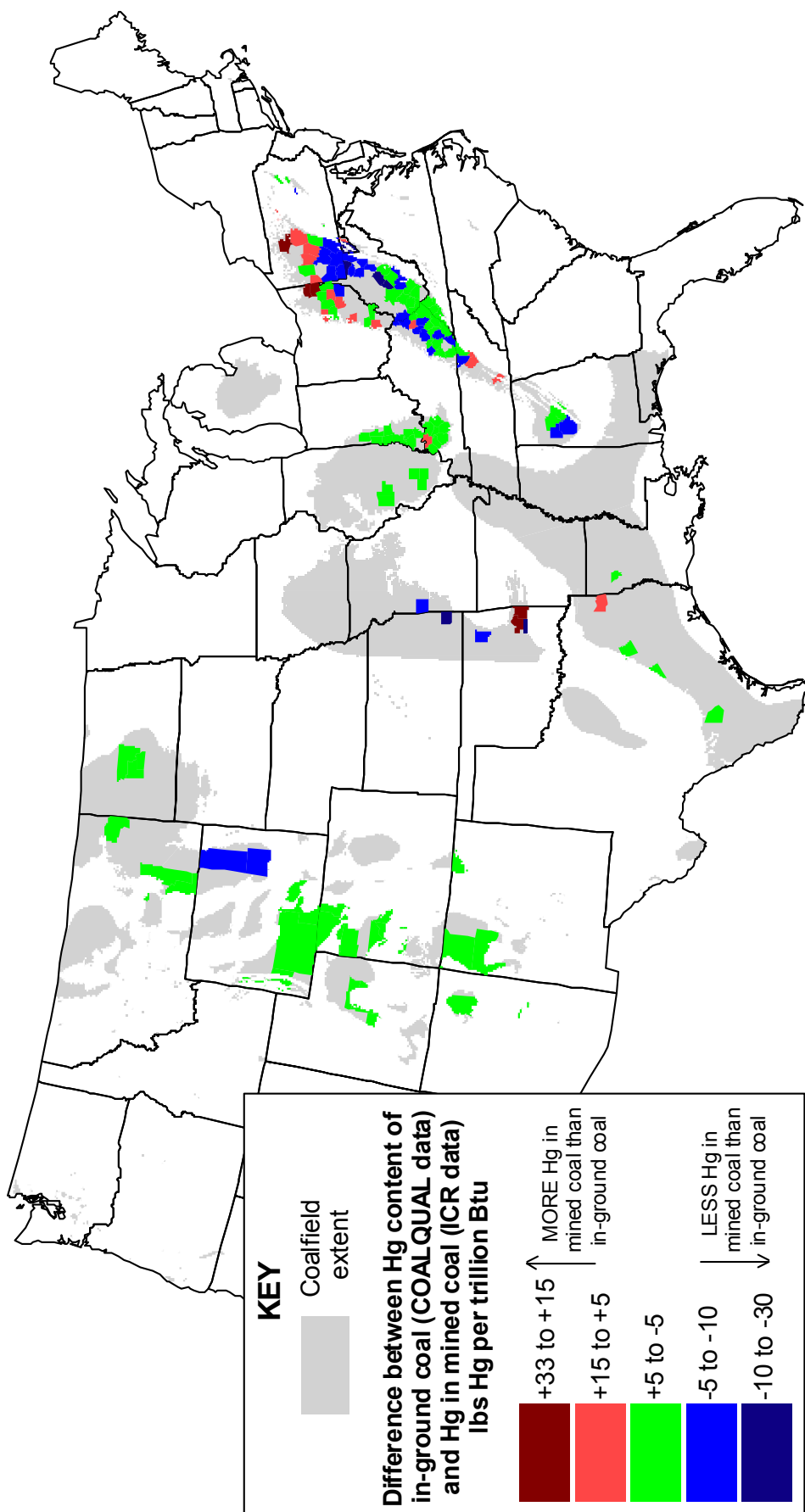
Figure 22. The difference between the mercury content of in-ground coal (COALQUAL data) and the mercury content of mined coal (ICR data) varies geographically (county-average COALQUAL Hg minus county-average ICR Hg, expressed as lbs Hg/10^{12} Btu).

KEY

Coalfield
extent

**Chlorine Content
ppm Cl (dry)**

< 200

200 - 600

600 - 1,000

1,000 - 1,500

1,500 - 2,500

> 2,500

Figure 23. Average chlorine content of coal delivered to U.S. power plants during 1999, by county-of-origin (calculated using selected ICR data).

Evaluation of Technology-Specific Hg Emissions

Figures 11 to 17 show the predicted county-average mercury emissions for coal burned in power plants classified by emission control technology. Note that these are draft figures and will likely be modified. About 70% of existing coal-fired utility boilers rely on either hot-side or cold-side ESP technology for emission control (Pavlish and others, 2003). Figures 11 and 13 show that eastern bituminous coal will rarely achieve the proposed MACT emission limit (2 lb $Hg/10^{12}$ Bt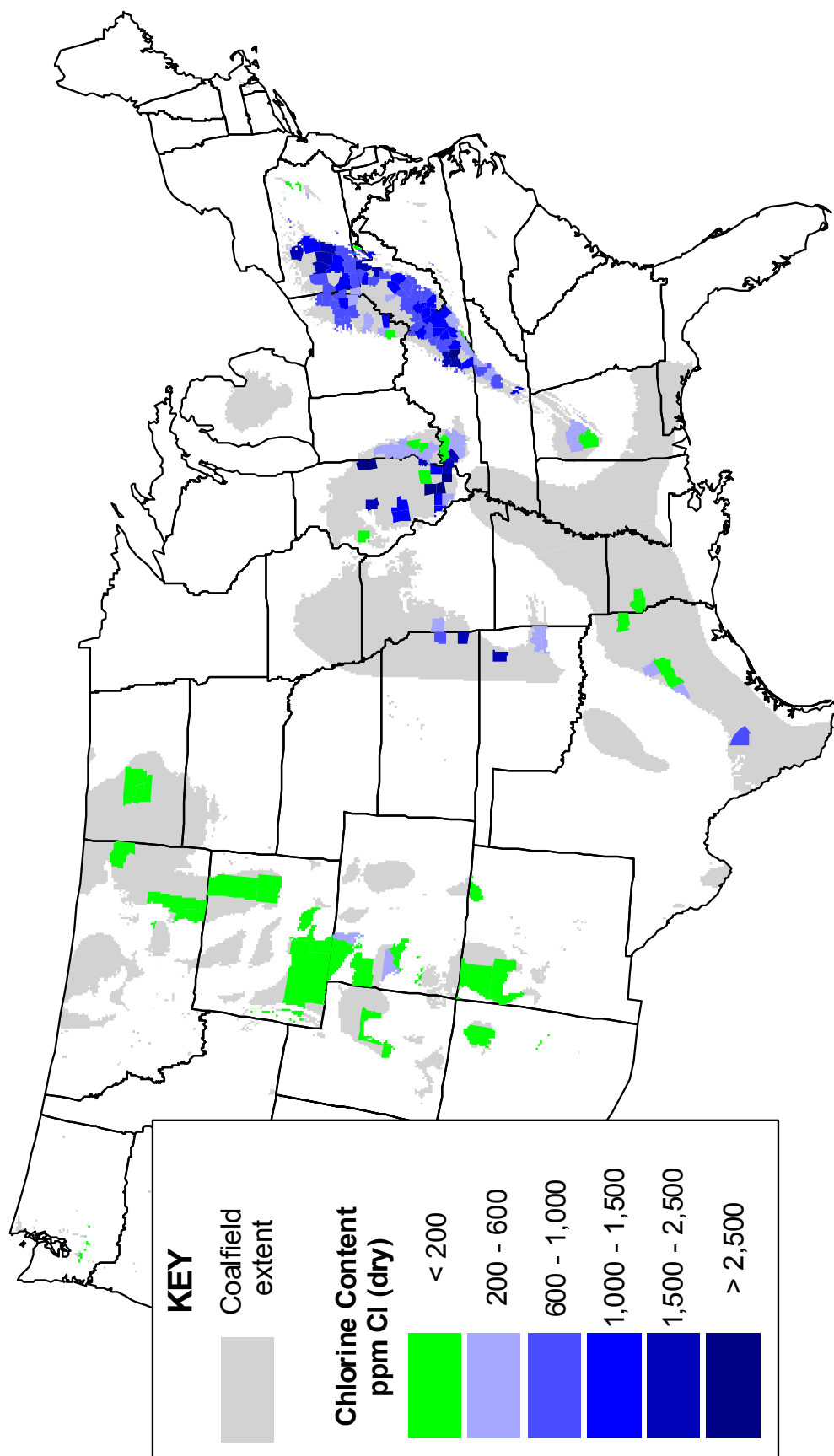u) for this substantial technology class. Likewise, no mercury compliance coals for these power plants are indicated for western bituminous coal (this includes 100% of Arizona and Utah production, 75% of Colorado production, and 38% of New Mexico production). Conversely, county-average values for most western subbituminous coal are below the proposed MACT limit (5.8 lb $Hg/10^{12}$ Btu). Given the higher MACT limit proposed for subbituminous coal compared to bituminous coal (table 1), switching to subbituminous coal may be an attractive compliance option for western power plants with ESP emission controls.

Considering the proposed MACT limit for plants burning lignite (9.2 lb $Hg/10^{12}$ Btu), the results for Northern Great Plains or Gulf Coast lignite burned in ESP equipped power plants are mixed. For example, in the Northern Great Plains, mercury compliance coal is indicated for Oliver Co., North Dakota and Richland Co., Montana, whereas the more significant coal production from McLean and Mercer Counties, North Dakota exceeds the proposed MACT limit.

About 12% of U.S. coal-fired utility boilers use FGD technology (Pavlish and others, 2003). Figures 12 and 14 show that the addition of FGD technology reduces mercury emissions, especially when combined with a cold-side ESP. Compared to ESP technology alone, there are more examples of mercury compliance coal for power plants equipped with FGD technology.

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However, despite better mercury capture using FGD, figures 12 and 14 show most bituminous coal-producing counties still exceed the proposed MACT limit if burned in plants using ESP-FGD emission controls.

Spray-dry-adsorption, fabric filter technology (SDA-FF) is used at about 4% of U.S. coal-fired utility boilers. Figure 15 indicates mercury compliance for bituminous coal from most counties when burned in power plants equipped with SDA-FF technology. However, the performance of SDA-FF technology is unlikely to be as good as indicated. Mercury emissions indicated by figure 15 are based on the county-average coal mercury and chlorine values, and the mercury emission rate was calculated using equation H listed in table 4. Figure 24 shows that the percent reduction of mercury emissions predicted by equation H (blue circles, SAIC 2003, model 1) is greater than what is predicted by other model equations. Consequently, the SAIC (2003) model is the most optimistic.

Although the different models for fabric filter technology shown in figure 24 clearly differ, they all indicate greater than 90% mercury capture above 1,200 ppm chlorine, as well as substantial sensitivity of predicted mercury capture when chlorine concentrations are below about 200 ppm. The sensitivity of the models below 200 ppm chlorine has special significance to western U.S. coal, given that the ICR chlorine assays are unreliable below this concentration (Nyberg, 2003), and that western U.S. coal commonly contains less than 200 ppm chlorine (figure 23).
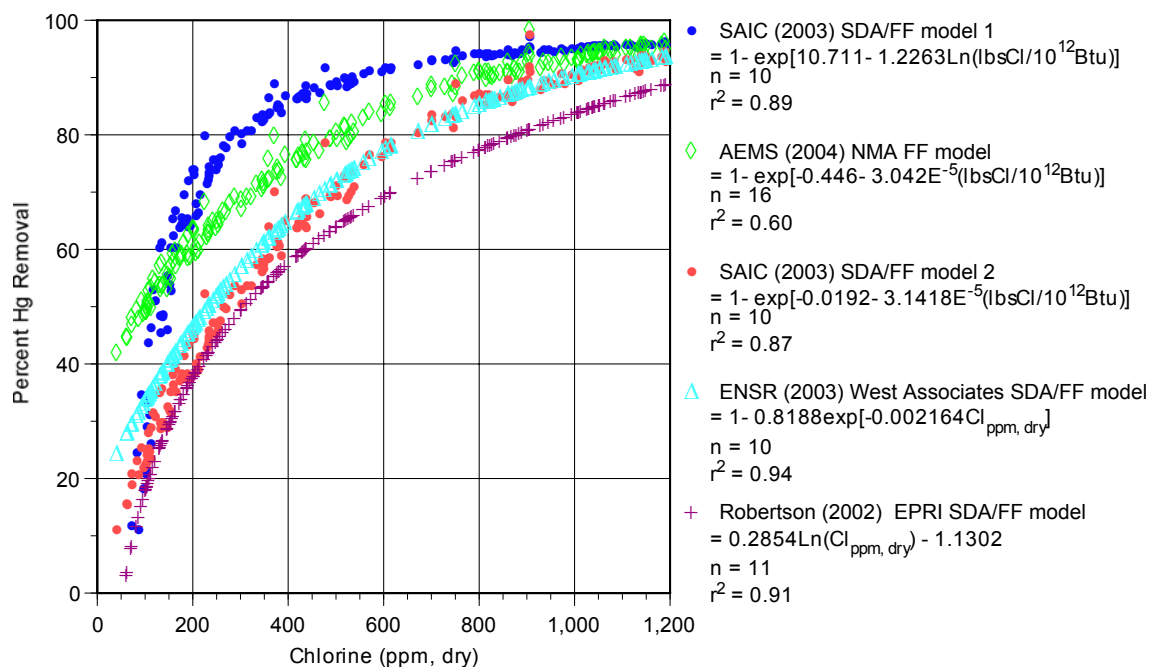
Figure 24. Five model equations predict increasing mercury capture by fabric filter emission controls with increasing coal chlorine concentration. Note that the SAIC (2003) model 1 equation was used to predict emissions in figures 15 and 17 of this report. Data points show county-average ICR chlorine values - and their variously predicted mercury capture efficiencies - for 184 U.S. counties; not shown are 46 U.S. counties where the average chlorine concentration is more than 1,200 ppm.

CONCLUSIONS

Draft maps showing potential sulfur, chlorine, and mercury emissions for U.S, coals by county-of origin, show the following:

- As generally known, high-sulfur coal is produced in the Northern Appalachian Region, as well as Eastern Interior, Western Interior, and Gulf Coast provinces, whereas low-sulfur coal is produced from the Central Appalachian Region and the western U.S.

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- High-mercury coal is produced in parts of Oklahoma, Texas, Ohio, Pennsylvania, Kentucky, Alabama, and Tennessee, whereas low-mercury coal is common in the western U.S., Eastern Interior Province, and the Central Appalachian Region.

- Coal from the Northern Appalachian Region (Ohio and parts of Pennsylvania) has notably high mercury concentrations, compared to other U.S. coal.

- Western U.S. coal typically contains less than 200 ppm chlorine, whereas eastern U.S. coal generally contains more than 600 ppm chlorine.

- Much subbituminous and some lignite coal should comply with the proposed MACT rule using existing technology. Bituminous compliance coal for power plants with ESP controls is rare. Plants equipped with FGD controls may find bituminous compliance coal in some western U.S. counties, the Eastern Interior Province, and the Central Appalachian Region. With notable exceptions (for example, certain counties in Ohio, Alabama, and the western U.S.), fabric filter technology may be an effective technology for most bituminous coal.

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